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SYNTHETIC SULPHIDE REPLACEMENT OF ORE MINERALS.[1]

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INTRODUCTION.

INVESTIGATION of the formation of metallic sulphides has been carried on since early in the nineteenth century.[2] This work has usually been done on more or less pure, finely divided, individual mineral varieties; generally in aqueous solutions with the addition of such gases as H_2S, SO_2, CO_2 etc. The object has been to determine:[3]

a. The chemical and physical conditions of precipitation relative to the genesis of the mineral.
b. The relative solubility (or dispersion?) of sulphide minerals in various types of solutions.
c. The differential electric potential of sulphide minerals.

Usually there has been no preconceived effort to induce metasomatic replacement of mixed sulphide minerals in massive specimens but at least two instances are given in the literature. Young

[1] Read at the annual meeting, Cordilleran Section, Geological Society of America, Univ. of Calif., Feb. 21, 22, 23, 1930.

[2] Regnault, *Annales de Chem. et de Phys.*, vol. 62, p. 378, 1836.

[3] For papers and bibliography of the subject see A. F. Rogers, *Sch. Min. Quar.*, vol. XXXII., pp. 298–304, 1911. F. F. Grout, ECON. GEOL., vol. VIII., p. 408, 1913. John D. Clark, Bull. Univ. of New Mex., 75, p. 142. Zies, Allen and Merwin. ECON. GEOL., vol. XI., pp. 408–503, 1916. S. W. Young, and Neil P. Moore, ECON. GEOL., vol. XI., pp. 349 and 574, 1916. R. G. Wells, U. S. G. S. Bulls. 548 and 609. Fred Foreman, ECON. GEOL., vol. XXIV., p. 811, 1929.

and Moore [4] obtained synthetic veinlets of chalcopyrite replacing bornite, and the same results are described by Zeis, Allen and Merwin.[4a]

Schwartz has recently done work with the so-called "graphic intergrowth" of bornite and chalcocite, using the direct application of dry heat (electric furnace).[5] At 175° C. he obtained incipient dispersion of bornite in chalcocite and at 250° C. he accomplished complete dispersion of bornite into solid solution in chalcocite. By slow cooling of the same specimen, after reheating to 250°, he produced a graphic design by the unmixing of the two minerals.

Some years ago (1914–1916) the writer noticed that "graphic" designs of bornite and chalcocite were ruined when specimens showing this relation were boiled in balsam. More recently (1928) the boiling point of the balsam has been determined as 150° C. thus lowering Schwartz's dispersion point, obtained in dry heat, by 25°.

The chemical investigations referred to have added greatly to our knowledge of the chemical reactions which are probably involved in enrichment processes but have yielded surprisingly few data on the actual replacement phenomena of ore minerals.

GEOCHEMICAL EXPERIMENTS.

The following described experiments were carried on with the hope of determining a further low point at which dispersion of bornite in chalcocite occurs as well as such replacement phenomena as might be induced under laboratory conditions.

If bornite dispersion in chalcocite can be induced at 175° C. under the influence of dry heat and at 150° C. in boiling balsam, a lower temperature might be expected if an excess of moisture were present. Water seemed a more natural environment than dry heat for metallic sulphide changes, especially where the minerals involved are admittedly of hydrothermal origin.

[4] J. J. Beeson, *T. A. I. M. E.*, vol. LIV., p. 398, 1917. S. W. Young and Neil P. Moore, ECON. GEOL., vol. XI., plate XXII. 1916.

[4a] *Op. cit.*, p. 479.

[5] G. M. Schwartz, ECON. GEOL., vol. XXIII., pp. 381–397, 1928.

Several bombs were made of 1 in. steel tubing. Polished ore fragments composed of pyrite, bornite, chalcocite and residual fragments of enargite were examined under the reflecting microscope and placed in the bombs. The bombs were filled one third with distilled water, sealed by means of thermo-welding and placed in gas furnaces which were maintained at temperatures of 175°, 150°, 125°, 100° C. respectively, for periods of from three to five days.

The specimens when examined after treatment all showed:

a. Complete dispersion of the smaller areas of bornite into the surrounding chalcocite. Partial dispersion of the larger bornite areas into the surrounding chalcocite.

b. Incipient and massive replacement of bornite by chalcopyrite where the bornite occurs in interstices between pyrite grains.

c. Incipient chalcocite replacement of pyrite.

Dispersion of bornite, in the presence of water, was thus obtained as low as 100° C.

Pyrex glass bombs were next used, immersed in a water bath maintained at a temperature of 90° to 100° C. No gases were introduced into the bombs, which were filled one third to one half with distilled water, sealed and placed in the bath, where they remained from seven to twenty days. Changes in the ore specimens were identical with those obtained at the higher temperatures. When cracked open, the thin end of the cooled bombs exploded outward thus indicating the formation of some gas during the course of the reactions involved.

Further experiments were made introducing copper sulphate, sulphur, sodium carbonate, and ferric sulphate into the water and results as listed below were obtained.

No chemical analyses of the solutions were made during these preliminary experiments. The solutions, where $CuSO_4$ was added to the distilled water, were a little below the saturation point at ordinary room temperatures.

TABLE I.

TABULATED DATA ON EXPERIMENTS.

Minerals in Specimen.*	Medium.	Temp. ° C.	Time in Days.	Reactions Obtained.
1, Py., Bo., Cc., (Eng., Kp.)	H_2O	90–100	7–16	Bo. → Cc. (solid solution) Bo. → Cp. (some Cov.) Py. → Cc. Kp. → Cc. + (Kp.?) Eng. → no change.
2, Py., Bo., Cc., Eng.	H_2O + $CuSO_4$	90–100	10	Bo. → Cc. (solid solution) Bo. → Cp. (minor amounts) Py. → Cc. (incipient)
3, Py., Bo., Cc., Eng.	H_2O + $CuSO_4$ + S.	90–100	6	Bo. → Cov. Cc. → Cov. Py. → no change. Eng. → no change.
4, Py., Bo., Cc., Eng.	H_2O + $CcSO_4$ + S + Na_2CO_3	90–100	6	Bo. → Cov. Cc. → Cov. Py., Eng. → no change.
5, Py., Bo., Cc.	H_2O + $CuSO_4$ + $FeSO_4$	90–100	7	Bo. → Cc. Formation of bright green triclinic crystals, basic copper sulphate, $3CuO.2SO_3.5H_2O$.†
6, Crystalline Sulphur.	H_2O + $CuSO_4$	90–100	7	S. → Cov. (massive, crystalline)
7, Sp., Ga., Qtz.	H_2O + $CuSO_4$	90–100	20	Sp. → Cc. + (Cov.) Ga. → Cov. Qtz. → no change.

* Qtz., quartz; Py., pyrite; Sp., sphalerite; Eng., enargite; Kp., klaprothite; Bo., bornite; Cov., covellite; Cc., chalcocite.

† The writer is indebted to Dr. A. F. Rogers for the optical and microchemical determinations of this compound. See footnote 6b.

MICROSCOPY.

The results of microscopic examination of the repolished surfaces of the treated ore fragments may be observed in the accompanying photomicrographs which are self-explanatory. They may be summarized as follows:

1, *a*. Complete synthetic dispersion of finely disseminated particles of bornite into chalcocite.

b. Nearly complete synthetic dispersion of larger masses of bornite into associated chalcocite; leaving faint blue "shadows" which mark the limits of earlier presence of pure bornite.

This phenomenon emphasizes the extreme instability of bornite and its tendency to give way to chalcocite, a fact which has long been evident to those who have pursued microscopic work on the sulphides of copper.

2, *a.* Synthetic metasomatic replacement of bornite by chalcopyrite and the development of "gash" veinlets, border zones of transition from bornite to chalcocite, sometimes accompanied with the formation of covellite.

b. The alteration of bornite to chalcopyrite is often accompanied by the replacement of adjacent pyrite by chalcocite. The manifestation of this replacement is such as to suggest that an electric couple, between pyrite and bornite, has been active, at least long enough to stimulate more general reactions. On a number of the specimens examined it was observed that incipient stages of chalcocite replacement of pyrite were evidenced by a succession of tiny patches of chalcocite on the pyrite. These patches were located in a line equi-distant from the edge of the pyrite grain on which they occurred and opposite areas of bornite which were being replaced by chalcopyrite.

The differential electric potential of sulphide minerals is discussed by Wells.[6] In the above observed occurrences an obvious source of the copper is the bornite, which suffers a reduction of copper during replacement by chalcopyrite, the chalcopyrite in turn receiving iron from the pyrite.

3. Synthetic replacement of pyrite by chalcocite in areas not necessarily contiguous with the deposition of chalcopyrite, but generally contiguous to bornite which is being dispersed into chalcocite. Solution of copper seems to be stimulated during or by the chalcocitization of bornite.

4. Synthetic replacement of klaprothite ($Cu_6Bi_4S_9$) [6a] by chalcocite accompanied either by a recrystallization of some klaprothite from solid solution in the chalcocite, or the development of a pseudo-eutectic design of residual klaprothite and chalcocite.

The above results were accomplished in water only at temperatures between 90°–100° C.

[6] *Op. cit.*

[6a] This seems to be the mineral generally referred to as klaprothite in the literature. It gives the microchemical reaction for bismuth.

5. In solutions of $CuSO_4$ in water the same type of alterations was observed with respect to the alteration of bornite to chalcocite, but no alteration of bornite to chalcopyrite was observed.
6. Synthetic formation of covellite as a metasomatic replacement of crystalline sulphur, the copper being supplied from a solution of $CuSO_4$ in water.
7. When crystalline sulphur was added to the $CuSO_4$ solution, bornite and chalcocite were both replaced synthetically by covellite. The synthetic metasomatism was accompanied by some shrinkage of volume, indicated by the development of shrinkage cracks in the covellite.
8. Synthetic metasomatic replacement of bornite and chalcocite by covellite when an excess of sulphur and $NaCO_3$ was added to the copper sulphate solution.
9. When $FeSO_4$ was added to the copper sulphate solution bornite was dispersed into the chalcocite. The solution precipitated out bright green triclinic crystals, identified by Dr. A. F. Rogers as $3CuO.2SO_3.5H_2O$, a basic copper sulphate.[6b]
10. Synthetic metasomatic replacement of sphalerite by chalcocite and covellite. This chalcocite replacement develops a pseudo-eutectic design in areas of partial replacement. Development of chalcocite appears to pass through the covellite stage; continued action of the copper-sulphate solutions furnishing an extra molecule of copper to form the cuprous sulphide. The internal crystallographic structure of the sphalerite exerts a control over the replacement design, certain parts being more resistant to replacement than others.
11. Synthetic metasomatic replacement of galena by covellite and the development of so-called "mutual boundaries." These "mutual boundaries" have often been interpreted as indicating contemporaneous deposition of the associated minerals but in this case the galena was collected in a mine and the covellite was made to replace it in the laboratory.

ANALYSIS OF RESULTS.

From the standpoint of metasomatic replacement the experiments described above have reproduced phenomena like and similar to those found in nature, *i.e.* replacements of one sulphide by another, mass for mass. They demonstrate that mixed sul-

[6b] This compound was produced and described by Posnjak and Tunell (*Amer. Jour. Sci.*, [5] vol. 18, p. 24, 1929).

phides in the presence of water only can generate their own conditions to bring about changes in these sulphides without addition of metals to the local system in which the changes are taking place. The reactions set up within the system generate the necessary mineralizers (of whatever nature they may be) which tend towards an intensification of the reactions themselves.

When $CuSO_4$ is introduced into a non-cupriferous system such as $ZnS + H_2O$ or $PbS + H_2O$ the more soluble salts of zinc and lead go into solution while the copper forms the insoluble cupric sulphide through a process of metasomatism.

If an excess of sulphur was added to the $CuSO_4$ solution, cupric sulphide resulted rather than cuprous sulphide as a replacement of bornite, and any cuprous sulphide already present in the ore was also converted to the cupric variety (covellite).

Although the chemical reactions involved in the above changes are probably more complicated than those generally recorded in the literature, the final results are quite plain under the microscope. Under laboratory conditions these changes have been brought about at temperatures below the boiling point of water, with a limited supply of oxygen, at low atmospheric pressure and during the course of a few days' time. It is probable that the same results would be obtained at lower temperatures and with longer time.

THE RÔLE OF BORNITE.[6c]

It will have been noted that bornite was extremely unstable under all conditions of the experiments. It must be concluded therefore that bornite is formed under some or all of the following conditions: above 175° C., under stronger deoxidizing conditions than those used in the writer's wet experiments or those made by others, and at greater atmospheric pressures. To the writer's knowledge there is no record in the literature of the synthetic formation of bornite except under conditions of a dry melt.[7]

[6c] The bornite here under discussion is not the obviously high-temperature bornite such as occurs at Ookiep and in certain other deposits described by Lindgren under "Bornite Deposits" in his Mineral Deposits, 1928, pp. 905–906 and classified by Graton and McLaughlin as "pneumotectic."

[7] J. D. Clark, U. S. Geol. Surv., Bull. 695, pp. 663–666. Also included references.

Within the writer's experience this type of bornite occurs well down in the sequence of sulphide deposition in hydrothermal deposits; in fact later than any of the unquestionably hypogene sulphides. In Butte, for example, it never replaces quartz as do pyrite, sphalerite, chalcopyrite, tetrahedrite and enargite but occurs either as a replacement of the earlier sulphides, calcite, or is deposited in open spaces.

Bornite is not a product of the shallower zones of downward secondary sulphide enrichment. It is, therefore, forced into a more or less uncertain "twilight" zone lying between undoubted hypogene deposition and the generally accepted zone of downward secondary sulphide enrichment. Rogers has suggested, for this type of early enrichment, the hypothesis of "upward secondary sulphide enrichment" to which he ascribes a hypogene origin.[8] The writer believes, however, that there is a simpler explanation for the occurrence of bornite in hydrothermal deposits (the mesothermal deposits of Lindgren).[9]

Bornite occurs only sparingly and as undoubted replacement remnants in chalcocite in the upper well defined zone of downward secondary sulphide enrichment. With increase in depth bornite generally occurs in relatively greater quantities with respect to chalcocite and its association with the latter mineral assumes a different aspect. At greater depth there appears an increasing tendency towards the development of "mutual boundaries" and "graphic" designs. These designs are interpreted by many to indicate contemporaneous deposition of the minerals so associated. The writer has for many years advanced opposition to this theory;[10] in general, study of a large number of specimens will reveal the so-called "graphic structure" or "mutual boundary" designs merging into structures of undoubted replacement origin. That such designs can be produced by the replacement of one sulphide by another has been shown in the present paper.

On the other hand Schwartz[11] has developed "graphic" struc-

[8] A. F. Rogers, ECON. GEOL., vol. VIII., pp. 781–794, 1913.
[9] Waldemar Lindgren, "Mineral Deposits," chapter XXVII., 1928.
[10] J. C. Ray, ECON. GEOL., vol. XI., pp. 179–185, 1916.
[11] *Op. cit.*

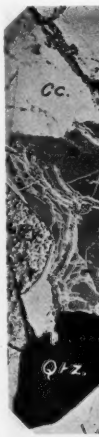
tures by slow cooling of chalcocite which contains bornite in solid solution. As pointed out, however, Schwartz's results were obtained under conditions of dry heat; a condition which is hardly compatible with natural conditions where moisture must surely be present at all stages of mineral deposition in hydrothermal deposits. It is possible that slow cooling under conditions of the writer's experiments will develop the "graphic" structure. Such experiments are being prepared at the present time and the results will be published at a later date.

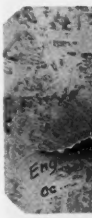
There is a third possible hypothesis which may account for the development of certain bornite-chalcocite "intergrowths." If massive bornite is undergoing replacement by chalcocite and the conditions of temperature, pressure and character of solutions are changed, so that some of the bornite is held in solid solution, then a gradual lowering of temperature might develop the much debated "graphic" design by recrystallization of the bornite. There would then be present two ages of bornite, a condition not at all incompatible with the observed natural occurrences, where massive bornite is undoubtedly undergoing replacement and the surrounding chalcocite contains patches of the bornite-chalcocite "graphic" pattern. It must be remembered, however, that "graphic" designs can usually be found which merge gradually into replacement structures.

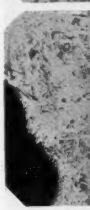
The genesis of the original (or "primary") bornite[12] has as yet not been agreed upon. If it be classified with the hydrothermal minerals, it is apparently the last to be deposited. If its lot is cast with the products of downward secondary sulphide enrichment, it is unquestionably the earliest. It is admittedly an unstable mineral. For this type of massive, deep-seated bornite, the writer advances the hypothesis of "deep-seated supergene alteration," and for the following reasons:

1. Bornite is deposited later than all minerals of undisputed hypogene origin.

[12] While the discussion is based specifically on the Butte ores, the writer believes that the principles here advanced have a general application to copper deposits of hydrothermal origin.







EXPLANATION OF FIGURES 1–5.

Qtz., quartz; Py., pyrite; Eng., enargite; Bo., bornite; Cc., chalcocite; bCc., blue chalcocite.

All specimens from Butte district, Montana.

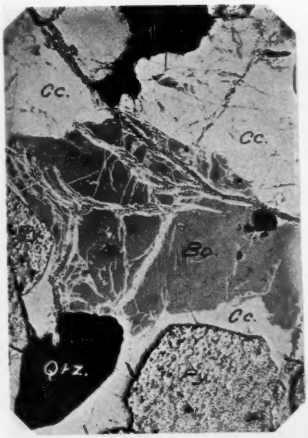
FIG. 1. R–764. × 30. Natural condition of ore fragment before laboratory treatment. Fragment from same specimen as those illustrated in 2 and 3.

FIG. 2. R–761. × 105. Synthetically induced partial dispersion of bornite into chalcocite. Light gray areas in chalcocite are blue chalcocite where complete dispersion has taken place. Gray patches, low relief, in pyrite show synthetic replacement of pyrite by chalcocite. Temp., 90°–100° C.; Time, 8 days; Solution, water only.

FIG. 3. R–806. × 37. Complete synthetically induced dispersion of bornite into chalcocite. Former area of bornite similar to those shown in Figs. 1 and 2 now indicated by shadowy outline of blue chalcocite. Gray spots on pyrite grain are synthetic replacement of the pyrite by chalcocite. Temp., 90°–100° C.; Time, 16 days; Solution, water only.

FIG. 4. R–809, *a* and *b*. × 60. Natural association of bornite and chalcocite. Same type of ore as that treated in Fig. 5 in which the "graphic" designs have been completely obliterated and the larger areas of bornite are shown as shadows of blue chalcocite.

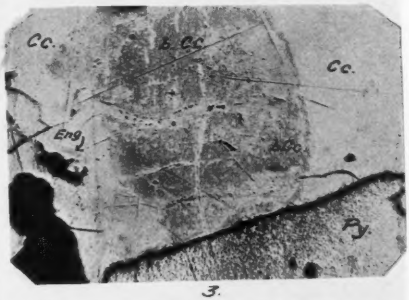
FIG. 5. R–762. × 45. This specimen originally had the same general distribution of bornite in chalcocite as that shown in 4. The finely distributed bornite has now entirely disappeared and the larger areas of bornite are indicated only by dim areas of blue chalcocite. Light gray spots on pyrite are synthetic chalcocite replacement. Temp., 90°–100° C.; Time, 16 days; Solution, water only.


Cc.
Cc.
Bo.
Cc.
Qtz.
Py.

1

Cc.
b Cc.
Cc.
Py.

2.


Cc.
b Cc.
Cc.
Eng.
Py.

3.

Py.
Cc
Bo.
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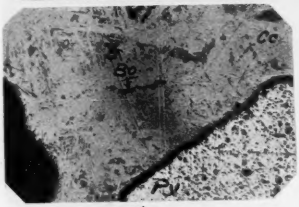
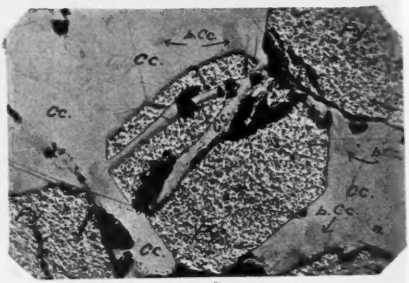
Eng.
Eng.
Cc.


Cc
Bo.
Py.

4.


Py.
Cc.
Cc.
Cc.
b Cc.
Cc.

5.

EXPLANATION OF FIGURES 6–10.

Py., pyrite; Tet., tetrahedrite; Bo., bornite; Cp. chalcopyrite; Cov., covellite.

All specimens from Butte district, Montana.

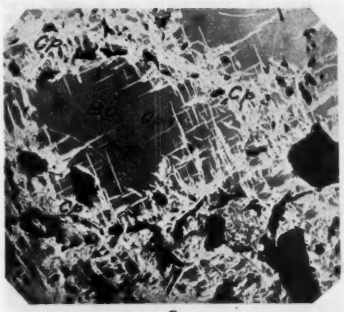
FIG. 6. R–733. ×65. Natural chalcopyrite developing "grid" structure in bornite. Some tetrahedrite also occurs along crystallographic directions of the bornite but is earlier than the chalcopyrite.

FIG. 7. R–743. ×90. Synthetic chalcopyrite developed along crystallographic directions in bornite. More prolonged heating develops massive replacement of the bornite. Temp., 90°–100° C.; Time, 8 days; Solution, water only.

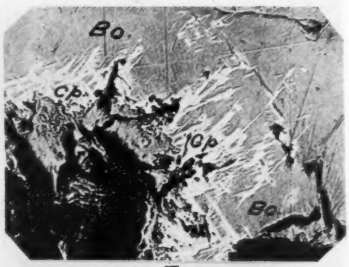
FIG. 8. R–805. ×90. Massive replacement of bornite by chalcopyrite and chalcocite, synthetically. Some covellite, also developed synthetically, seems to be an intermediate stage between chalcopyrite and chalcocite. Compare with covellite, natural, as in Fig. 10. Temp., 90°–100° C.; Time, 16 days; Solution, water only.

FIG. 9. R–804. ×90. Synthetic replacement of bornite by massive chalcopyrite and chalcocite. The bornite originally occupied the interstitial spaces between pyrite grains and was traversed by veinlets of chalcocite similar to those shown in Fig. 1. Presumably prolonged heating would induce further alteration of the chalcopyrite to chalcocite. Temp., 90°–100° C.; Time, 16 days; Solution, water only.

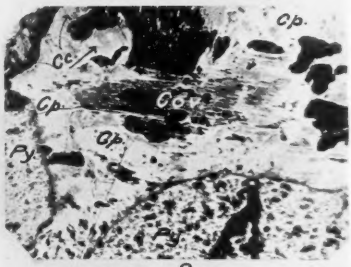
FIG. 10. R–698. ×210. Natural occurrence of covellite and chalcopyrite. Here, however, study of many specimens leads to the conclusion that chalcopyrite is replacing covellite. Note similarity of occurrence to the synthetic relations shown in Fig. 8.


Cp.
Cp.
Bo.
Cp.

6.


Bo.
Cp.
Cp.
Bo.

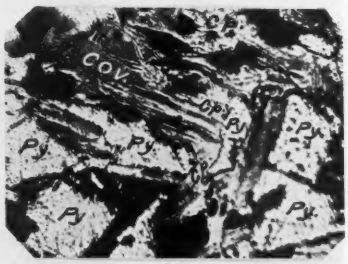
7.


Cp.
Cc.
Cp.
Cc.
Cp.
Py.
Py.

8.

Py.
Cc.
Cp.
Py.
Cp.
Cc.

9.


Cp.
Cc.
Cp.
Py.
Py.
Py.
Py.
Py.

10.

EXPLANATION OF FIGURES 11–14.

Qtz., quartz; Py., pyrite; Kp., klaprothite; Ga., galena; Bo., bornite; Cov., covellite; Cc., chalcocite.

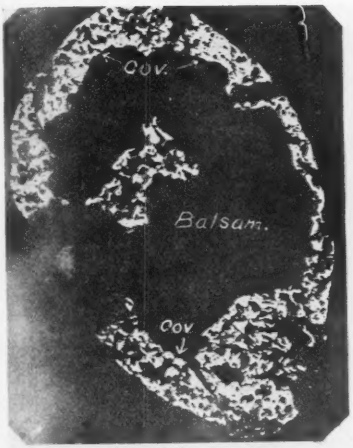
All specimens from Butte district, Montana.

FIG. 11. R–803. × 60. Synthetic covellite produced by replacement of sulphur fragments in $CuSO_4$ solution. The fragments were cemented in balsam to permit of polishing. The centers of many of the fragments were unreplaced sulphur, the sulphur going into solution in the heated balsam. Temp., 90°–100° C.; Time, 7 days; Solution, water + $CuSO_4$.

FIG. 12. R–758. × 45. Synthetic covellite as metasomatic replacement of bornite and chalcocite. Some shrinkage of volume is indicated by the development of cracks (black) in the covellite. Temp. 90°–100° C.; Time, 6 days; Solution, water + $CuSO_4$ + S.

FIG. 13. R–742. × 75. Klaprothite ($Cu_6Bi_4S_9$) showing incipient synthetic replacement by chalcocite. The veinlets existed as tiny fractures in the klaprothite before the specimen was treated in the laboratory. The white specks in the replacing chalcocite are either residual Kp. or are a second generation thrown down with the crystallizing chalcocite. See also replacement structure shown in Fig. 17. Temp., 90°–100° C.; Time, 8 days; Solution, water only.

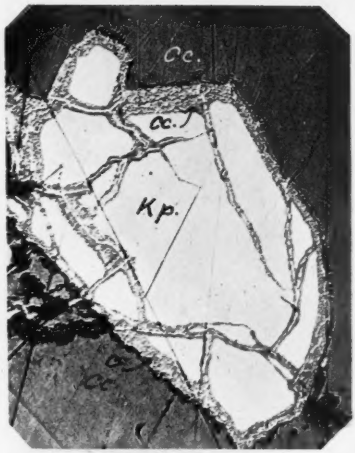
FIG. 14. R–756. × 60. Same ore fragment as in Fig. 17. Synthetic covellite metasomatically replacing galena, so-called "mutual boundaries" between galena and covellite which is so frequently referred to as indicative of "simultaneous deposition." Having been produced in the laboratory the covellite is obviously later than the galena. Note that the covellite replacement has been volume for volume and no shrinkage cracks are developed as is the case in Fig. 12.


Cov.
Balsam.
Cov

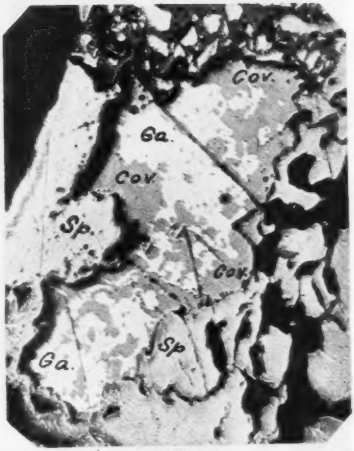
11.

Py.
Py
Cov
Qtz.
Py
Cov

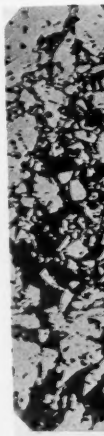
12.

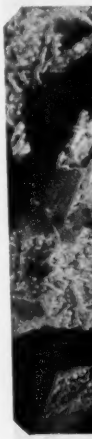

Cc.
Cc.
Kp.
Cc
Cc

13.


Cov.
Ga.
Cov
Sp.
Cov
Ga.
Sp

14.





EXPLANATION OF FIGURES 15–18.

Q., quartz; Sp., sphalerite; Cov., covellite; Cc. chalcocite.

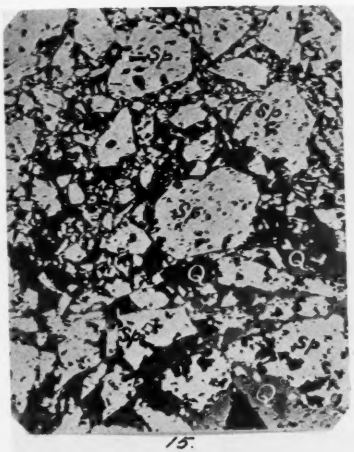
All specimens from Butte district, Montana.

FIG. 15. R–182. × 35. Natural occurrence of brecciated sphalerite cemented in quartz. Fragments of this specimen used in Figs. 16, 17, and 18 illustrated below.

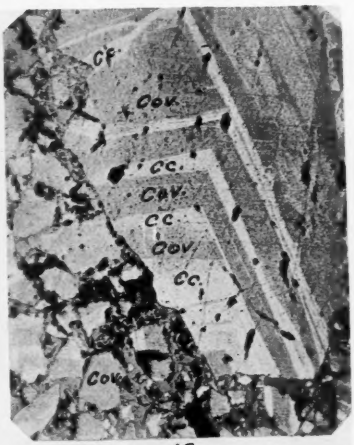
FIG. 16. R–755. × 120. Same type of ore as that illustrated in 15. Incipient synthetic replacement of sphalerite by covellite and chalcocite. The design shown is determined by the crystalline structure of the sphalerite. Covellite seems to be induced where there is a concentration of iron in the earlier sphalerite; chalcocite directly replaces the sphalerite where there is less iron present. Temp., 90°–100° C.; Time, 10 days; Solution, water + $CuSO_4$.

FIG. 17. R–760. × 185. Synthetic metasomatic replacement of sphalerite by covellite and chalcocite. Note the pseudo-graphic design developed between residual sphalerite and the replacing chalcocite. High relief, dark gray, covellite; high relief, light gray, sphalerite; Low relief, light gray, chalcocite. Temp., 90°–100° C.; Time, 20 days; Solution, water + $CuSO_4$.

FIG. 18. R–810. × 65. Same experimental fragment as shown in Fig. 17, showing differential resistance of sphalerite to replacement. Synthetic chalcocite replacement is more complete along certain crystallographic directions of the sphalerite. Pseudo-graphic design is developed in the sphalerite by partial replacement. Same conditions of treatment as in Fig. 17. The specimen has been polished down until more of the residual sphalerite is exposed. High relief, sphalerite; Low relief, synthetic chalcocite.


Sp
Sp
Sp
Q
Q
Sp
Sp
Q

15.


Cp
Cov.
Cc.
Cov.
Cc
Cov
Cc.
Cov

16.

Sp
Sp
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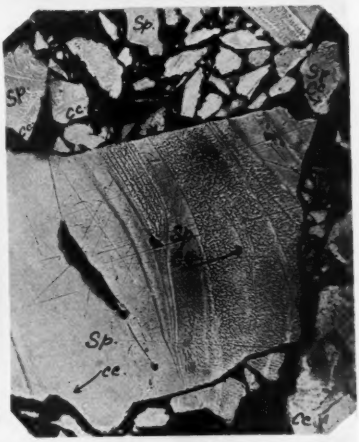
Sp
Cc.
Cc.

17.


Sp
Sp
Cc.
Sp
Sp.
Sp.
Cc.
Cc.

18.

2. It occurs always as a replacement of earlier sulphides, or deposited in open spaces.
3. It does not replace quartz but it does replace calcite, a gangue mineral which is itself deposited later than quartz, and later than sulphide minerals of undoubted hypogene origin.
4. Deposition of bornite marks a decided break in the introduction of metals into the deposit.
   a. Up to the time of the deposition of bornite the deposit has received additions of pyrite, chalcopyrite, sphalerite, galena, tetrahedrite, enargite and many rarer minerals.
   b. With the introduction of bornite begins the elimination of all earlier metals with the exception of copper, *i.e.*, iron, zinc, lead, antimony, arsenic, the rarer metals and sulphur.
5. Bornite can be traced back into the zone of intensive downward secondary sulphide enrichment in decreasingly small amounts and through all stages of replacement by chalcocite, chalcocite being the final point of copper enrichment and elimination of other metals.
6. Bornite is formed under conditions that seem to postulate high atmospheric pressure rather than high temperature (as applied to hydrothermal processes). Being formed with high pressure as the controlling factor might account for the extreme unstability of the mineral.
7. Through the many geological periods which have passed since the deposition of our great ore deposits, structural conditions must at times have made it possible for meteoric waters to penetrate to relatively great depths. This would be particularly true in regions of deep veins where diastrophism would tend to bring about fault block readjustment. A classic example is the Butte district, Montana.

   At depths of ten to twelve thousand feet below the earth's surface, the normal rise in temperature with depth should give a rock temperature of 100° C.[13] Add to this the probability of residual heat from the slowly cooling igneous ore bringer and hydrostatic pressure up to several hundred atmospheres, then a condition is created which has potent possibilities for the reworking of ore minerals. Chemical reactions once started, the process would continue until gradual cooling, erosion and lightened pressure no longer would make possible the formation of an unstable mineral such as bornite.

[13] Waldemar Lindgren, *op. cit.*, p. 598.

Downward secondary sulphide enrichment accomplishes very considerable concentration of copper which has been leached from the zone of oxidation. On the other hand the deep-seated supergene alteration outlined above accomplishes rather a reworking of such ores as are present with comparatively small addition of copper. The writer has computed that the copper present in bornite and chalcocite of the deeper levels of the Butte district is an increase of only about 10% over that originally present in the earlier hypogene ores composed largely of enargite and tetrahedrite.[14] Gradual diffusion of leached copper from above supplies that element for replacement of earlier noncupriferous sulphides.

ACKNOWLEDGMENTS.

The writer is indebted to Dr. S. W. Young for laboratory facilities and advice on the procedure of the chemical work of the experiments, and to Professor C. F. Tolman for critical discussion of the subject matter of the paper.

[14] Unpublished thesis, Stanford University.

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LIMESTONE OIL RESERVOIRS OF THE NORTHEASTERN UNITED STATES AND OF ONTARIO, CANADA.[1]

A. N. MURRAY.

I

INTRODUCTION.

Most of the oil fields of the northeastern United States and Ontario, Canada, were developed between 1860 and 1890, at a time when little attention was given to oil-well data and drill cuttings, and many of the pools were opened and operated by individuals and small groups of men who had no knowledge of geology and no appreciation of its worth. Owing to these conditions the data which would have thrown much light on the petroleum geology of these important fields have been lost.

Since the presence of a porous reservoir is essential to an oil field, the problem of the origin of porosity becomes important, and this paper is an attempt to show how the porosity was developed within the limestones of the area under consideration. The material upon which this investigation is based was obtained from the field, from the various State Geological Surveys, from companies interested in the area and from the literature.

The writer wishes to express his appreciation of the assistance rendered by Dr. W. V. Howard, under whose direction the work was carried on, as well as the counsel given him by Drs. W.

Submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Geology in the Graduate School of the University of Illinois, 1928.

[1] This paper contains preliminary results obtained in an investigation of limestones as reservoir rocks, listed as Project No. 23 of the American Petroleum Institute Research. Financial assistance in this work was received from a research fund donated by the Universal Oil Products Company to the American Petroleum Institute. This fund is being administered by the Institute with the coöperation of the Central Petroleum Committee of the National Research Council.

S. Bayley, T. E. Savage, and A. C. Bevan, all of the Department of Geology, University of Illinois.

POROSITY.

The term porosity when applied to rocks, is used to designate the amount of open space per unit volume. The statement that a limestone has a porosity of fifteen per cent may not mean that the rock is an oil reservoir, since cavities or groups of cavities may be sealed in such a way that they have no connection with neighboring cavities. In order that a limestone may become a reservoir for oil, the cavities must be inter-communicating so that when the rock is pierced by the drill, the oil which may have collected in it may flow or drain out by passing from opening to opening.

Porosity may thus be subdivided into continuous porosity, suited to the accumulation of oil, and discontinuous porosity, unfavorable for accumulation. It is to be noted, of course, that indigenous oil may be contained in cavities which are not connected. Oil, when in limestone with this type of porosity, is not available under present methods of production.

The Niagara limestone at the quarry of the Monon Stone Company at Monon, Indiana, possesses discontinuous porosity. In this limestone some of the cavities, which are mainly the molds of fossils, are filled with liquid brown oil, whereas adjacent ones are not. In this case the distribution of oil-filled and of empty cavities shows clearly that the oil entered the limestone from some other source. Thus at one time the porosity must have been continuous through large sections of the limestone. It was rendered discontinuous by the deposition of calcite crystals in some of the openings, and the presence of empty cavities lined with these crystals shows that the deposition of calcite began before oil entered the rock. The precipitation of calcite continued along with the infiltration of oil with the result that many crystals contain inclusions of liquid oil (Fig. 1). Eventually the passages leading from the source became completely closed and both the oil and water circulation ceased.

The Devonian limestone in the quarries of the St. Mary's Cement Company, St. Mary's, Ontario, also shows discontinuous

FIG. 1. Calcite crystals containing inclusions of oil, taken from cavities in Niagara limestone from quarries of Monon Stone Company, Monon, Indiana.

porosity associated with oil (Fig. 2). Because of the lack of passage ways between the cavities, commercial production cannot be obtained from these rocks.

PRIMARY POROSITY IN LIMESTONES.

The term primary porosity is used to designate the openings in limestones which exist by virtue of the process by which, and

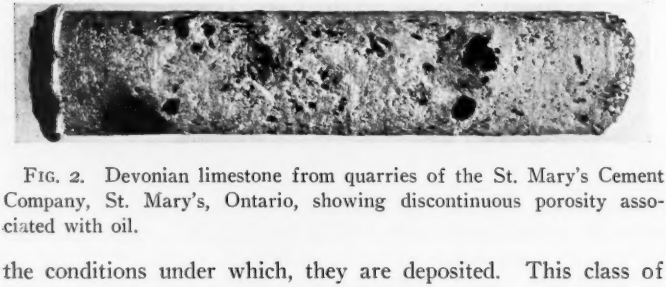

FIG. 2. Devonian limestone from quarries of the St. Mary's Cement Company, St. Mary's, Ontario, showing discontinuous porosity associated with oil.

the conditions under which, they are deposited. This class of porosity may be possessed by all limestones and varies directly

with the uniformity of size of grain and inversely with the degree of induration and packing. It tends to be of the discontinuous rather than of the continuous type because of the filling in of the interstices between the grains or fragments with finer material and cement. It is therefore concluded that limestone with primary porosity would not form an important reservoir for oil. The field evidence and the result of prospecting for oil support this conclusion.

SECONDARY POROSITY IN LIMESTONES.

The term secondary porosity is applied to the openings in a limestone developed after the rock has been deposited and indurated. By far the larger number of limestone reservoirs have porosity of this type. In practically all the oil fields of the northeastern United States the porosity has been caused by the action of chemical agencies on limestone after deposition and induration.

No evidence has been found which proves that the development of porosity by the solvent action of carbonic acid in sea water cannot take place, but there seems to be no reason for considering that this action is important in producing reservoirs, and other causes of the development of porosity must be considered.

Dolomitization and its Relation to Porosity.—The presence of oil in porous dolomitic limestones has led many to believe that the process of dolomitization has been responsible for the development of the openings in which the oil occurs. The theory that the dolomitization of a limestone produces porosity appeared in the literature early in the nineteenth century. The evidence, if such there was, which gave it support seems to have been lost while the theory itself lingers. The writer was unable to find in the literature any criterion by which porosity by dolomitization can be distinguished from primary porosity or porosity developed by leaching and solution. Many dolomites are described in which the porosity is supposed to have been caused by this process, but no observer, as far as can be determined, has ever advanced any evidence in favor of this conclusion.

If the replacement were one of molecule for molecule, a loss

of 12.3 per cent.[2] in volume would result. Igneous rocks, on loss of volume, due to cooling, do not develop porosity but joints and cracks; the same is true of sediments on dehydration and of mud on drying. It seems, then, that dolomitization, molecule for molecule, which would result in a decrease in volume, would pro-

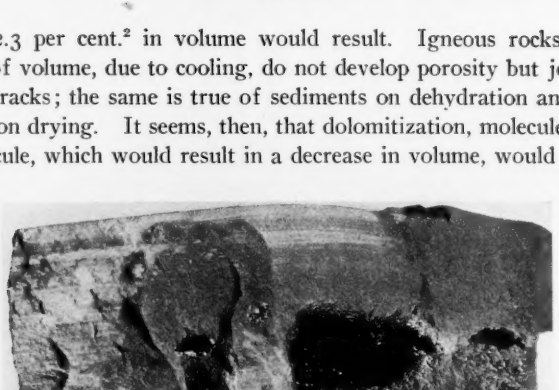

FIG. 3. Cavity cutting across bedding. Silurian dolomite, Kokomo, Indiana.

duce fracturing and perhaps a decrease in thickness of the beds, but not porosity.

Beaumont,[3] who was the first to calculate the amount of shrinkage which would be produced by the dolomitization of calcium carbonate, molecule for molecule, believed that he could explain the cavernous nature of the dolomites of Tyrol on this basis. A later porosity determination of 12.9 per cent. for a single sample of dolomite from the Alps by Morlet[4] seemed to confirm Beaumont's idea.

[2] Van Hise, C. R., "A Treatise on Metamorphism," U. S. Geol. Surv. Mon. 47, p. 379.

[3] Cited by F. M. Van Tuyl, "The Origin of Dolomite," Iowa Geol. Surv., Bull. 25, p. 286, 1914.

[4] Cited by F. M. Van Tuyl, *op. cit.*, p. 286.

The presence of large numbers of porous beds between dense beds of dolomite and dense mottlings of dolomite in limestones show that dolomitization is not a molecular replacement but usually a volume replacement. Cavities which cut across laminated dolomite without deformation, as they do in the Silurian dolomites at Kokomo, Indiana (Fig. 3) tend to show that the cavities were not produced by shrinkage.

Moreover, if it is assumed that the porosity of dolomites is developed by shrinkage, then dolomitization by volume replacement must change to that by molecular replacement within very short distances. In the Irvine oil field, Estill County, Kentucky, the reservoir rock is a porous dolomite in the upper parts of the Onondaga limestone, but this same formation in a railroad cut northwest of the town of Irvine, not over two miles from the field, is so dense that it has the appearance of a quartzite, and even within the Irvine field itself, wells have been drilled which have found the producing horizon compact and without oil.[5] In the oil-producing areas of southwestern Ontario, the presence of the Marcellus shale above the Onondaga is considered a "bad sign" by the drillers.[6] Since the presence of oil in this formation seems to be dependent on the development of porosity, and since the Onondaga horizons that produce oil are dolomitic, whether or not they are porous enough to become a reservoir, it is not easily understood why the presence of Marcellus shale should induce volume replacement with little or no porosity, and its absence permit molecular replacement with the development of porosity sufficient for an oil reservoir.

Judging from the lack of field evidence, the peculiar and erratic occurrence of porosity in dolomites, and the way in which other rocks are affected by shrinkage, the conclusion seems justified that little, if any, of the porosity in dolomitic limestones is developed by decrease in volume produced by molecular replacement. It is therefore necessary to turn to some other process for the development of reservoirs.

[5] St. Clair, Stuart, *Trans. Amer. Inst. Min. and Met. Eng.*, vol. 65, pp. 165–175, 1921.

[6] Bell, R. N., *Proc. Roy. Soc. Can.*, vol. 5, p. 107, 1887.

POST-EMERGENT POROSITY.

Secondary post-emergent porosity may be developed by the solvent action of mineral salts, carbonic acid and organic acids on calcareous limestones or on magnesian or dolomitic limestones. In the former case porosity is developed as a result of partial solution of the rock and in the latter by differential solution whereby the less soluble compounds are left behind as a porous mass which is capable of forming an excellent reservoir for petroleum.

During the elevation of sedimentary rocks above sea level stresses are set up in the rocks by compression, tension and torsion. These form fractures and joints as well as larger structural features and there are also many incipient fractures produced, which are not visible until the rock has been exposed to weathering, when they are emphasized by chemical erosion.

When sediments are elevated, the joints and fractures provide access for meteoric waters carrying mineral salts, acids and carbon dioxide to the strata below.

Although limestones tend to be more soluble in solutions of salts than in pure water, the solubility of limestones in such dilute solutions as those which make up the ground water is very slight. Carbonic acid is much more effective and the acids formed by bacterial decomposition of organic matter have been shown to be most efficient in dissolving calcium carbonate.[7]

Solutions entering the rocks travel downward until the water table is reached, where they begin to travel laterally, or nearly so, towards some surface drainage system. The depth to which the water sinks before it begins its horizontal movement is influenced by the relief, the presence or absence of impervious beds, the amount of water falling on the surface and the permeability of the rock. In some areas, particularly those of concentrated rainfall or marked seasonal changes, there are wide fluctuations in the depth of the water table.

In order that calcium carbonate may be dissolved by ground water, circulation sufficiently rapid to remove the calcium bicar-

[7] Murray, A. N., and Love, W. W., "Action of Organic Acids on Limestone," *Bull. Amer. Assoc. Petrol. Geol.*, vol. 13, No. 11, pp. 1467–1475, Nov. 1929.

bonate is necessary. Thus, in limestone regions where there are no impervious strata to prevent percolation of the waters, porosity or cavernous conditions may be developed from the surface almost to the level of the drainage of that area, but not farther. Water with some carbon dioxide will reach the water table, and as it travels laterally, it will spend the rest of its carbon dioxide as it passes out to the surface drainage system. As the water table is lowered the waters are permitted to percolate through longer distances, and if the depth of the water table is lowered sufficiently the carbon dioxide may be neutralized before reaching it.

Below the water table, no solution takes place, because here the solutions are saturated with calcium carbonate and bicarbonate. Stagnation prevents the bringing in of additional carbonic acid and the removal of the bicarbonates already formed. A slight decrease in the hydrostatic head or an increase in the temperature will produce supersaturation in these stagnant solutions: this will result in cavity filling and cementation.

Porosity Developed by Leaching.—When two substances with different solubilities are attacked, the one with the greater solubility may be almost completely dissolved before the one with the lesser solubility is affected. If two such substances form a rock, the removal of the more soluble leaves the less soluble honeycombed with cavities.

Calcite is much more soluble than dolomite in carbonic acid under normal conditions.[8] Specimens which contain calcite and dolomite may be readily etched by acids which dissolve the calcite leaving the latter in relief.

The writer subjected small fragments of calcite, dolomite and magnesite to the action of a 2 per cent. solution of acetic acid. The calcite effervesced rapidly, the dolomite gave off a few bubbles and the magnesite an occasional bubble. At the end of twenty hours about two-thirds of the calcite had disappeared, and at the end of forty-eight hours it was completely dissolved. The dolomite took four weeks to dissolve and the magnesite re-

[8] Hardman, E. T., *Proc. Roy. Irish Acad.*, 2d Sess., vol. 2; pp. 711–715, 1875–77. See also Van Tuyl, F. M., *op. cit.*, p. 316.

mained, after three months of exposure to the acid, with the edges of the rhombs sharp, the faces smooth and apparently with no loss in volume.

These results have been repeatedly given by other experimenters, some of whom place the solubility of calcite at least twenty-five times as great as that of dolomite. The latter is soluble, but slowly, in dilute acid, and on dissolving yields a solution which is very close to the dolomite ratio.

Field Evidence of Leaching.—If calcite is leached from dolomitic limestone by meteoric waters, there must be considerable evidence of the fact by solutions percolating through these rocks and by the waters drained from areas underlain by dolomitic limestone. There should also be evidence in the physical and mineralogical character of the undissolved rock, both on the present surface and below old buried erosion surfaces.

Analyses of the Onondaga limestone of Ohio show that the rock has a calcium magnesium ratio of 1.7 : 1 whereas waters from this series in Ohio have calcium magnesium ratios of from 2.0 : 1 to 8.0 : 1 with an average of 4.1 : 1.[9] Similarly the Niagara limestones of Ohio with a calcium magnesium ratio of 1.72 : 1 are associated with waters whose calcium magnesium ratios range from 1.92 : 1 to 4.0 : 1.[10] Travertine deposits formed by waters from the Onondaga limestone are of almost pure calcium carbonate.[11] These data show that there is a tendency toward the retention of magnesium carbonate in the rock and a leaching of the calcite.

The data provided by surface streams are less satisfactory in that few analyses of the waters of streams which drain areas underlain only by dolomitic limestones are available. Moreover, such analyses would show the results of leaching a rock which had already been subjected to weathering for a prolonged period. The following ratios were calculated from a list given by Steidtmann.[12] As far as possible a selection was made of those rivers

[9] Orton, E., U. S. Geol. Surv., 19th. Ann. Rept., Pt. 4; pp. 637–717, 1897–1898.
[10] Steidtmann, E., *Jour. of Geol.*, vol. 19, p. 415, 1911.
[11] Orton, E., *op. cit.*, p. 681.
[12] Steidtmann, E., *op. cit.*, p. 416.

which drain an area underlain by one dolomitic formation. Some descriptive material has been added by the present writer.

River.	Location.	Formation.	Ca.	Mg.	Period of Observation.
Fox River	Elgin, Ill.	Niagara dolomite.	1.70	1	1 year
Kankakee River	Kankakee, Ill.	Niagara dolomite.	2.76	1	do.
Rock River	Rockford, Ill.	Galena-Platteville mostly dolomitic limestone.	1.80	1	do.
White River	Azalia, Ind.	Silurian, mostly dolomitic limestone.	2.65	1	do.
Wabash River	Logansport, Ind.	do.	2.34	1	do.
Miami River	Dayton, Ohio	Niagaran, mostly dolomitic limestone.	2.45	1	do.

The abnormally low ratio for the water from the Rock River is undoubtedly due to the weathered condition of the Galena dolomite from which the larger part of the calcite has already been leached. Whether or not this explanation can be applied in the case of the Fox River is not known, for there seems to be no available information regarding the weathering of the Niagara limestone in that area and it was not visited by the writer.

As in the case of the travertine deposits referred to above, cave deposits, both stalactitic and stalagmitic, which are deposited from waters which have passed through the overlying limestones, throw considerable light on the nature of the material dissolved by downward percolating water. These are almost invariably much richer in calcium carbonate than the rocks in which the caves appear.[13] Moreover the veinlets which commonly traverse deformed limestones are mostly of calcite, cavities in dolomitic limestones lined with calcite are common, and there is abundant evidence in the presence of fossil casts and molds of the removal of calcite from dolomitic limestone.

Steidtmann has already noted the great decrease in the calcium-magnesium ratio in weathered limestone[14] and Bevan[15]

[13] *Cf.* Hardman, E. T., *op. cit.*, pp. 718–719, and Van Tuyl, F. M., *op. cit.*, p. 387.
[14] *Op. cit.*, p. 394.
[15] Personal communication.

has found in the Oregon Quadrangle, Illinois, that the Galena limestone has been leached locally to such an extent that cavities are developed which are partly filled with sand composed of what appear to be rhombs of dolomite together with some calcite. The cavities, which are not more than two inches in diameter, are produced by the crumbling of the rock after the calcite has been leached by meteoric waters. Leaching of this type is more marked on the hills than in the valleys.

In drilling into the Monroe formation of Michigan, dolomitic sands are encountered which flow into the holes like thin mud. Smith [16] considers that such sands and oozes may be the residuum of beds which have been almost completely removed by solution.

In addition to many porous dolomitic horizons lying below unconformities in North America, it is interesting to note that Hardman [17] described beds of cavernous dolomitic limestone interstratified with blue fossiliferous limestone from County Tyrone, Ireland. He estimates that half of the original rock is wanting and thinks that the cavities are undoubtedly the work of differential solution.

Summary.—Experiments on calcite and dolomite have shown that calcite is many times more soluble in weak acids than dolomite, and analyses of ground water and of streams which have their source in areas underlain by dolomitic limestone show that magnesium is being retained in the rocks whereas calcium is being removed. The residues of the rocks are therefore much higher in magnesium and lower in calcium than the original rocks and more porous, many having porosities greater than could possibly have been rendered by dolomitization. Since organic acids and carbonic acid operate most efficiently on the land surface where circulation has been established, it is concluded that the porosity of dolomitic limestones is largely produced by leaching action while they are at or near the surface of a land mass. If this is true, limestones below unconformable surfaces should be more porous than similar limestones differently situated.

[16] Smith, R. A., Michigan Geol. and Biol. Surv., Pub. 14, Geol. Ser. 11, pp. 206–208, 1912.

[17] *Proc. Roy. Irish Acad.*, 2d Ser., Science 2, p. 723, 1875–77.

UNCONFORMITIES AND THEIR RELATIONS TO RESERVOIR ROCKS.

The oil-producing dolomitic limestones of northeastern United States and Ontario are all of Paleozoic age. During this era the land masses in Ontario, Ohio, Indiana, Illinois, and Kentucky were, in general, of low relief. Streams were sluggish and slopes gentle, so that rain falling on the surface was not delivered rapidly to the sea. Carbonic and organic acids were carried slowly down into the rocks by meteoric waters and, on reaching the water table, slowly migrated laterally to the surface drainage system. Such a condition would be ideal for the development of porosity by solution and leaching wherever a limestone or dolomitic limestone was exposed at the surface.

Development of Porous Strata Seemingly not Connected with Unconformity.—In a series of sediments where crystalline dolomitic limestones alternate with dense calcareous ones, solution takes place more rapidly in the former than in the latter. Waters percolating down through the former dissolve the calcite, giving rise to porosity of the continuous type, whereas in the latter, solution takes place along joints and fissures, producing caves and underground streams. J. M. Weller [18] states that one of the requisites for extensive development of underground drainage by means of sinks and caves is a thick series of massive, compact and rather pure limestones, which lie high above the main lines of surface drainage.

Porosity might easily be developed in a dolomitic limestone underlying a compact calcareous limestone, provided a part of the former lay above the water table and the latter rock was traversed by fractures or joints. Water entering such a series from the surface would pass downward by way of the fractures into the coarse-grained bed where differential solution would take place with the consequent development of porosity.

If a porous stratum becomes an oil reservoir, undoubtedly the unconformity, which might lie many feet above the porous stratum, would not be immediately associated with the develop-

[18] Geology of Edmonson County, Kentucky Geol. Surv., Ser. 6, vol. 28, p. 48, 1927.

ment of the reservoir. In cases of this kind, however, the erosion interval represented by the unconformity is the immediate cause of the existence of the porosity.

Proof that ground water is active under a considerable cover is found in the Dundee limestone of Michigan. The "Bottomless" sinkhole,[19] Sec. 36, T. 33N., R. S. E., has a depth of 50 feet (estimated) to the water, which has a similar or greater depth. The upper 20 feet of the walls of the sinkhole consist of limestone of the Long Lake series, underlain by the Bell shales, which are about 80 feet thick; the Dundee limestone lies below these. Here solution beneath 80 feet of shale has taken place to such an extent that sink holes have been produced. In this particular case solution has probably been caused by water supplied from the surface through cracks in the shale.

Development of More than One Porous Zone Below an Unconformity.—It is reasonable to expect that solution will take place at the ground water table more readily than above it because of the concentration of the waters which have percolated through the rocks or descended by joints and fissures. A stationary or almost stationary water table will give rise to a zone with greater porosity than the overlying rock, but if for some reason the surface drainage system were lowered, the increased gradient given the water table by virtue of this lowering would cause it to migrate downward until it approached the level of the surrounding surface drainage, where it would remain fairly stationary, probably producing another porous zone.

Evidence supporting the development of cavernous and porous limestones at different depths by the variable rate of down-cutting of the surface streams is found in the different levels existing in caves.[20]

The Green and Nolin rivers in Edmonson County evidently existed for some time at an elevation of around 700 feet, developing a fairly large-sized, flat and mature valley topography. During this time the water table, which was fairly stationary, developed the first level in the caves of the region. Rejuvenation

[19] Grabau, A. W., Mich. Geol. Surv. Ann. Rep., 1901, p. 190.
[20] Weller, J. M., *op. cit.*, p. 37, 1927.

caused the rivers to cut rapidly downward to an elevation of around 600 feet. The level of the water table migrated downward to keep pace with the river and did not permit the production of any marked passages or channels during this period, but as soon as the stream came to a temporary rest at about 600 feet, the ground water again developed a system of underground streams at or near the water table. By a repetition of this procedure the third level was produced.

If, on the other hand, the surface drainage system cuts downward at a fairly constant rate, such porosity as is developed in the limestone should be quite uniform, unless the process is impeded by shaly partings and dense calcareous limestone beds.

Relief of Erosion Surface and its Relation to Porosity.—Limestones, in order to be rendered porous by the action of either carbonic or organic acids on an erosion surface, must lie above the level of the surface drainage system. Streams do not have to be numerous or the elevations high to establish the circulation which must exist to produce porosity.

Small, irregular, and irregularly distributed noses in the oil fields of Wood, Sandusky, and Hancock Counties, Ohio, strongly suggest low hills which were buried in the muds of the Utica sea, and later tilted by the Cincinnati uplift. The irregular distribution of oil wells on small noses in many limestone fields can be readily understood if explained in this way. Large structures with considerable closure could not be so interpreted, since there is no indication that there was high relief in the Mississippi Valley at any time during the Paleozoic era.

Effects of Submergence on Porous Limestones and Topographic Forms.—Rocks exposed at the surface will be affected by wave erosion when covered by an advancing sea. The amount of erosion accomplished depends upon the rate of submergence, the character of the rock, and the amount of the relief of the land surface. It is evident that topographic forms composed of soft rock, friable or easily broken and corroded, such as leached or cavernous limestone, will suffer greater modification under these circumstances than will forms composed of denser, harder varieties of the same kind of rock. The more rapidly

a sea advances over a land mass, the less will be the chance of great modification of the topography by cliff erosion. It would be hard to conceive of an advancing sea which had no effect on the topographic forms over which it was advancing. Fenneman [21] states that no supposition is allowable which would engulf an island so promptly that the waves could not etch its descending slopes. The greater the relief of the surface over which the sea is advancing, the greater will be the truncation and paring down of the hills. Kindle,[22] in studying the unconformity at the base of the Chattanooga shale in Kentucky, has observed a considerable number of irregularities. He found at Rileys, Kentucky, a residual clay directly overlying the Onondaga and partly filling the depression.

The waters of a sea advancing over a porous limestone area immediately fill all the pores with water containing the salts common to sea water. The waves, in the process of sorting and grading the sediments, wash material into the open pores, sealing the sea water in the limestone. Kindle's figure 25 represents a solution cavity filled with black shale of the overlying Chattanooga.[23] This cavity lay at least 8 feet below the erosion surface at the top of the Devonian limestone.

FISSURES AND JOINTS AS RESERVOIRS.

Many wells have been drilled into limestone reservoirs in which the tools have been known to drop from a few inches to several feet. This may be caused by the striking of a solution cavity or a fissure. The dropping of tools is very common in the Ohio field, but here it is definitely known that the rock is of a cavernous nature by the character of the fragments of reservoir rock blown from wells on shooting. The wells do not seem to behave in the manner expected of fissure wells.

At Terre Haute, Indiana,[24] a well drilled in 1888 came in a gusher. Much drilling followed, but only four wells struck oil,

[21] *Geol. Soc. Amer. Bull.* 16, p. 207, 1905.
[22] Kindle, E. M., *Amer. Jour. Sci.* (4), vol. 33, p. 123, 1912.
[23] *Idem.*, p. 126.
[24] Scoville, H. F., Ind. Dept. G. and N. R., Ann. Rept. 21, p. 527, 1896.

and all of these were within a very small area. Wells drilled very close to the four producers failed to find oil. The second producing well obtained in this pool was pumping 60 bbls. of oil per day after eight years of production. There is a belief that the oil was conducted from another field by a passage, but it seems to the writer to be more logical to assume that it was probably fissure production, the oil being supplied from a reservoir below, which would have been tapped by the other wells if drilled deep enough.

In the Spring Creek area, Overton County, Tennessee,[25] the oil production is from the cherty limestone of the Fort Payne formation of Mississippian age. The production of the first well drilled was completely stopped by the drilling of another well to the same horizon. It was probably this peculiar behavior of the wells which led Munn[26] to state that the oil of the Spring Creek field apparently comes entirely from crevices in the Fort Payne formation. To the writer this seems the only logical conclusion when the behavior of the wells is considered.

THE LIMESTONE RESERVOIRS OF THE NORTHEASTERN UNITED STATES AND ONTARIO.

Trenton Limestone.—The Trenton limestone of Ordovician age is the principal producer of gas in Indiana and forms the main reservoir of oil in the Lima-Indiana oil fields. Of late years a producing horizon has been found within the Trenton, but the great bulk of the oil is obtained from near the top of the formation. Ulrich[27] shows that there was not only an unconformity between the Trenton and the overlying Utica, but there were also local unconformities within the Trenton itself. Deposits of late Trenton age are wholly wanting in the Mississippi Valley to the north of the Ohio River,[28] so that the occurrence

[25] Munn, M. J., Tenn. Geol. Surv. Bull. 2, Extract E, pp. 6–7, 1911.

[26] *Idem*, p. 8.

[27] Ulrich, E. O., "Revision of the Paleozoic system," *Bull. Geol. Soc. Amer.*, vol. 22, p. 416, 1911.

[28] Ulrich, E. O., "The Ordovician—Silurian Boundary," Cong. Geol. Int., C. R. XII. Sess., p. 643, 1913.

of oil in the Kimmswick and Galena limestones of Illinois is also probably the result of accumulation of oil in a limestone which was made porous during late Trenton time.

In Ontario and Michigan, the Trenton limestone is also locally petroliferous and it is believed that conditions of unconformity prevailed during late Trenton time in this region as well as in the area to the south and west.

Guelph Formation.—The Guelph produces both oil and gas in Ontario, and Williams [29] ascribes the variable thickness of the formation to post-Guelph and pre-Camillus erosion.

Other Silurian Reservoirs.—In Kentucky much of the production ascribed to the Onondaga limestone (Corniferous) is actually from the Niagara limestone of Silurian age.[30] The fact that the Onondaga rests directly upon the Niagara in Kentucky indicates the presence of an unconformity above the latter formation.

In Illinois, the productive Niagara limestone is overlain unconformably by the Sweetland Creek shale of Devonian age.[31]

Devonian Limestones.—In Ontario, limestones of Onondaga age are productive only where they are not separated from the overlying Delaware limestones by shales of Marcellus age.[32]

The Hamilton (formerly Onondaga or Corniferous) limestone forms the most important reservoir of oil in Kentucky where it is overlain unconformably by the Chattanooga shales,[33] and this unconformity extends northwestward into Indiana and Illinois, where Devonian limestones are also productive.

In Michigan, according to Newcombe,[34] there is a strong probability that the Dundee limestone (Devonian) has been removed by erosion over much of Michigan and that the Monroe limestones which were exposed as a result of this erosion are reservoir rocks in the Michigan fields.

[29] Williams, M. Y., Geol. Surv. Can., Memoir 111, p. 72, 1919.

[30] St. Clair, Stuart, *Bull. Amer. Assoc. Pet. Geol.*, vol. 6, p. 30, 1922. Shaw, E. W., U. S. Geol. Surv. Bull. 661, pp. 157–158, 1917.

[31] Collingwood, D. M., Ill. Geol. Surv., Rept. of Invest. No. 1, p. 10, 1924.

[32] Bell, R. N., *Proc. Roy. Soc. Can.*, vol. 5, p. 108, 1887.

[33] Kindle, E. M., *op. cit.*, pp. 120–136.

[34] Newcombe, R. B., "Middle Devonian Unconformity in Michigan," *Bull. Geol. Soc. Amer.*, 1930 (from manuscript).

Traverse Formation.—The Traverse formation in Michigan produces from two main horizons. Of these the first is in the top of the Alpena-Petosky member, where it is 200–210 feet thick as opposed to a thickness of 350 feet in the type locality, and from the top of the Long Lake limestone below an anhydrite bed of local extent.[35] Lane[36] considers that there "is good reason to suppose that during late Traverse there was some emergence." The thinning of the Alpena-Petosky member would seem to bear this out. By analogy with the relations between anhydrite and limestone in many parts of the world, a local unconformity is suspected above the Long Lake limestone.[37]

Lower Mississippian Limestones.—In Illinois, oil is found in a number of limestones of Lower Mississippian age, including the Salem and Ste. Genevieve limestones. Mylius considers that porosity was developed in these limestones as a result of erosion.[38]

Summary and Conclusions.—It has been pointed out that on theoretical grounds the most effective, if not the only, way in which sufficient porosity can be developed in limestone to provide a reservoir of commercial importance is as a result of solution above a water table. The porous limestone reservoirs in the northeastern United States and Ontario all have been shown to have been subjected to erosion before the time of deposition of the formation now overlying them, with the possible exception of the limestones of Traverse age in Michigan. It is therefore suggested that erosion caused the development of porosity in the case of all these limestone reservoirs.

[35] Newcombe, R. B., *op. cit.*

[36] Lane, A. C., Notes on the Geologic Section of Michigan, Pt. 2, *Jour. Geol.*, vol. 18, p. 417, 1920.

[37] *Cf.* Pohl, Erwin R., The Middle Devonian Traverse group of Rocks in Michigan, *Proc., U. S. Nat. Museum*, vol. 76, Art. 14, pp. 1–34, 1929.

[38] Mylius, L. A., Ill. State Geol. Surv. Bull. 54, 1928.

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A QUALITATIVE AND QUANTITATIVE DETERMINATION OF THE ORES OF COBALT, ONT.[1]

PART I.

ELLIS THOMSON.

CONTENTS.

Part I.

Introduction.
Methods of Analysis: Development of New Etching Reagents.
Geology.
Origin and Description of Specimens Used in Investigation, Mining Corporation, Nipissing and O'Brien Mines.
General Conclusions from Detailed Descriptions of Specimens from Mining Corporation, Nipissing and O'Brien Mines.

Part II.

Relations of the Metallic Minerals to the Diabase Sill and the Enclosing Country Rocks.
Description of Additional Specimens from Other Parts of the District.
Rare Mineral Occurrences.
Ore Genesis.
Summary.

INTRODUCTION AND PRELIMINARY INVESTIGATIONS.

A STUDY of the minerals contained in the ores from this district is an interesting but difficult problem. Much interesting data has been compiled from a study of the ores of this district by Anderson, Campbell and Knight, Emmons, Miller, Palmer and

[1] Submitted as partial fulfilment of the requirements of the degree of Doctor of Philosophy, Harvard University. The work on which the paper is based was done in the Mineralogical Laboratory, University of Toronto, under the direction of Professor L. C. Graton, Harvard University.

Bastin, Van Hise, Whitehead, and others.[2] Most of this information was of a geological or physico-chemical character and little attempt was made to differentiate the hard white metallic arsenides and sulpharsenides which are the common associates of the native silver. Although it is true that such minerals as cobaltite, arsenopyrite, and rammelsbergite were cited as mineralogical curiosities, it was assumed that the great bulk of these hard white minerals was made up of the familiar smaltite-chloanthite mixture. A study of silver ores from the Keeley Mine, South Lorraine, by the writer, carried out in 1925,[3] seemed to cast some doubt on this assumption and the present investigation was started partly with a view to confirming this suspicion or otherwise. Such constituents as native silver, native bismuth, niccolite and breithauptite are, of course, easy of identification under the microscope; but there remains a residue of hard white minerals, of about the same physical properties, that have many more points of similarity than of difference. The latter will be enumerated more particularly later but are mentioned here as the first problem to solve before any great progress in mineral determination can be expected. The improved polarizing attachment for the mineragraphic microscope, which has been developed in the last few years, is of material assistance in this regard. It was felt, however, that extra microchemical data would have to be assembled for the final solution of this problem. While some progress had been made already in this direction by Baumhauer [4] and Vollhardt,[5] by Bastin [6] and by Ellsworth [7] in his pioneer effort

[2] Anderson, R., *Eng. & Min. Jour.*, vol. 88, p. 786.
Campbell, W., and Knight, C. W., *Eng. & Min. Jour.*, June 9, 1906; ECON. GEOL., vol. 1, pp. 767–776.
Emmons, S. F., *Min & Sci. Press*, San Francisco, Cal., 1911, pp. 140–156.
Miller, W. G., *Ont. Bur. of Mines*, vol. 19, Pt. 2, pp. 1–133.
Palmer, C., and Bastin, E. S., ECON. GEOL., vol. 8, pp. 140–170.
Van Hise, C. R., *Can. Min. Inst. Jour.*, vol. 10, pp. 45–61.
Whitehead, W. T., ECON. GEOL., vol. 15, pp. 103–135.

[3] Thomson, E. Univ. of Toronto Studies, Geol. Series, No. 20, pp. 54–58.

[4] Baumhauer, A., *Zeit. für Kryst.*, 12, pp. 18–33.

[5] Vollhardt, G., *Zeit. für Kryst.*, 14, p. 407.

[6] Bastin, E. S., ECON. GEOL., vol. 20, pp. 8–10.

[7] Ellsworth, H. V., *Ont. Bur. of Mines*, vol. 25, Pt. I., pp. 200–243.

on the ores from Cobalt, Ont., and by the writer [8] in his study of the ores from the Keeley Mine in South Lorraine, Ont., as well as by Todd [9] in his determination of the ores from Gowganda, Ont., it was felt that a better differentiation of the mineral constituents of this group could be effected with the aid of new microchemical reagents.

METHODS OF ANALYSIS.

It might be well at this stage to give the names, compositions and crystal habits of the hard white metallic minerals of this group before proceeding to the microchemical part of the subject. They are as follows:

Name.	Composition.	Crystal System.
Cobaltite	$CoAsS$	Cubic
Gersdorffite	$NiAsS$	Cubic
Arsenopyrite	$FeAsS$	Orthorhombic
Skutterudite	$CoAs_3$	Cubic
Smaltite	$CoAs_2$	Cubic
Safflorite	$CoAs_2$	Orthorhombic
Chloanthite	$NiAs_2$	Cubic
Rammelsbergite	$NiAs_2$	Orthorhombic
Loellingite	$FeAs_2$	Orthorhombic

To this list might be added nickel skutterudite, the nickel cobalt triarsenide which is known but has not as yet been observed in the silver ores of Northern Ontario. In his work on the ores from the Keeley Mine,[10] the writer found that a varying degree of susceptibility to chemical reagents was shown by the different minerals of this group. It was found, for instance, that rammelsbergite was the most easily attacked by dilute nitric acid, but both skutterudite and cobaltite were unaffected by this reagent. With this characteristic in view it was decided to try first the effect produced by different strengths of the four standard reagents of Davy-Farnham,[11] leaving the reagent on the polished surface a standard time of one minute. Nitric acid being the

[8] *Op. cit.*

[9] Todd, E. W., *Ont. Dept. of Mines*, vol. 35, Pt. 3, pp. 62–78.

[10] *Op. cit.*

[11] Davy-Farnham, "Microscopic Examination of the Ore Minerals."

only one of these four to give a decided reaction on all but cobaltite and skutterudite, dilutions of this reagent were experimented with first. It was found that a mixture of three parts of concentrated acid and seven of water left all but rammelsbergite and safflorite unaffected. Of these two the rammelsbergite was etched a light brown color while safflorite showed a very light brown or gray. A saturated solution of ferric chloride on the other hand etched only rammelsbergite, staining it a dark brown and loellingite, a light brown or yellow, leaving the other seven minerals unaffected. A strong solution of potassium cyanide left all these minerals unaffected. As the two supplementary reagents, potassium hydroxide and mercuric chloride, though in saturated solutions, left most of these minerals unaffected, experimentation with these reagents seemed to be quite unprofitable and they were passed over. Many other reagents were made up and their etching reactions tested, for the most part with unsatisfactory results. In the great majority of instances their reactions were either too general or else too feeble to be of any practical value in the differentiation of these minerals. The following list of reagents that were tried and found ineffective, is appended for the benefit of those investigators who may be pursuing research along the same lines:

Sodium sulphide.
Silver nitrate and nitric acid mixture.
Iodic acid.
Glacial acetic acid.
Sulphur chloride.
Monochloracetic acid.
Dichloracetic acid.
Trichloracetic acid.
Ammonium persulphate.
Potassium iodide.
Acetyl chloride.
Ferric ammonium sulphate.
Perchloric acid.
Arsenious chloride in HCl.
Benzoyl chloride.
Antimonous chloride in HCl.

Potassium dichromate in H_2SO_4; in HNO_3; in HCl.
Potassium ferrocyanide.
Copper sulphate.
Picric acid in H_2O.
Stannous chloride.
Potassium zinc iodide.
Potassium mercuric iodide.
Potassium cadmium iodide.
Conc. H_2SO_4.
Conc. HCl.
HNO_3 (4 in 10).
Phosphoric acid.
Barium nitrate—nitric acid mixture.
Potassium dichromate in sulphuric and nitric acid mixture.
Aqua regia (1 in 10); (2 in 10); (3 in 10); (4 in 10); (5 in 10).

As will be seen from the above list, some of these reagents were tried also in different dilutions but without success.

A saturated solution of potassium permanganate on the other hand was found to etch gersdorffite a light to dark brown in most sections, leaving the other eight minerals unaffected, which represented the best effort at differentiation obtained with any reagent.

The principle of saturating a solution with one or other of the three metals concerned, Fe, Co, or Ni, was next tried, using aqua regia as the solvent. It was hoped that if the solution became saturated for Fe, it might act on the Co and Ni minerals and leave the Fe minerals unattacked. The following solutions were experimented with, only one being at all successful:

1. Aqua regia (40 cc.) and Ni (5.5 gms.).
2. Aqua regia (50 cc.) and Ni (3.25 gms.).
3. Aqua regia (45 cc.) and Co (3 gms.).
4. Aqua regia (50 cc.) and Fe (5 gms.).
5. Aqua regia (80 cc.) and Co (3 gms.) and Ni (3 gms.).
6. Aqua regia (165 cc.) and Co (3 gms.) and Ni (3 gms.) Water.
   70% 30%
7. Aqua regia (95 cc.) and Co (3 gms.)
8. Aqua regia (95 cc.) and Co (3 gms.) Water.
   70% 30%
9. Aqua regia (50 cc.) and Fe (5 gms.) Water.
   30% 70%

Of the above solutions, No. 4 separated five of the group of minerals from the other four, giving etching reactions for loellingite, rammelsbergite, gersdorffite, smaltite, and chloanthite, and negative results on safflorite, arsenopyrite, skutterudite, and cobaltite. No 9, a dilution of No. 4 with water, reduced the number of these positive reactions by one, gersdorffite being unaffected by this reagent.

Variation of the time for etching, using aqua regia as the reagent, from ten seconds to one minute, also failed to produce satisfactory results.

Returning then to the idea of saturation, experiments were next carried out, using the actual mineral compounds, as the solutes and aqua regia again as the solvent. Since these nine minerals might be conveniently divided into three classes, according to the metal present, viz:

1. Iron class { loellingite ($FeAs_2$); arsenopyrite (FeAsS).
2. Cobalt class { smaltite ($CoAs_2$); safflorite ($CoAs_2$); skutterudite ($CoAs_2$); cobaltite (CoAsS).
3. Nickel class { chloanthite ($NiAs_2$); rammelsbergite ($NiAs_2$); gersdorffite (NiAsS).

it seemed probable that an attempt to saturate the solution with the mineral compounds, CoAsS, NiAsS, FeAsS, might lead to slightly better results than when using the metals above. As comparatively pure arsenopyrite (FeAsS) was readily available, a solution of this material in aqua regia was made as nearly saturated as possible, the solution being hastened by heating and the final product being filtered through an asbestos filter to free it from the undissolved material. This solution was found to be more satisfactory as it separated rammelsbergite and chloanthite from the other seven minerals. In dealing with a similar solution of cobaltite (CoAsS) it was found impossible to obtain any considerable quantity of pure material in the natural form, so a

mixture of Co, As, and S in the proper proportions was substituted. This was not as good a differentiating agent as the iron compound but did separate loellingite, gersdorffite, rammelsbergite, and chloanthite, by positive reactions, from the other five with negative reactions. Still another of these saturated solutions was experimented with, using both Co and Ni with As and S, made up as a substitute for the compound (Co, Ni) AsS, with a view to segregating the two iron compounds from the other minerals. This solution was made in a similar way to the other two, using 7.3 gms. of Co, 7.3 gms. of Ni, 9 gms. of As, 4 gms. of S, and 80 cc. of aqua regia. It appeared to be not quite saturated for Co, as it attacked the safflorite with a differential reaction and etched smaltite and chloanthite to a pale brown color, leaving the other six minerals unaffected.

The results obtained from the use of these saturated or semi-saturated solutions seem to bear out the principle that it is possible to separate one of the three classes from the other two and, while those so far obtained are by no means complete, they give promise of better things in the future when more detailed investigation may open up new and successful channels of attack not only for such compounds as are here studied but for many other opaque minerals.

In order that some measure of the susceptibility of the nine minerals under discussion might be obtained, a table has been prepared showing the positive reactions obtained for twenty-four of the reagents used. The latter were chosen entirely at random and included the last three saturated solutions, as well as many of the unsuccessful special reagents. This table is given herewith:

TABLE I.

Name of Mineral.	No. of Postive Reactions.
Rammelsbergite	19
Gersdorffite	16
Chloanthite	15
Smaltite	14
Loellingite	13
Safflorite	9
Arsenopyrite	7
Skutterudite	1
Cobaltite	0

TABLE I.

Minerals.	Conc. HNO_3.	1 – 1 HNO_3.	3 in 10 HNO_3.	Pot. Perm.	$FeCl_3$ (Sat.)	Aqua Regia + Fe.	Aqua Regia + Fe + H_2O.	Aqua Regia + FeAsS.	Aqua Regia + CoAsS.	Aqua Regia + (Co, Ni)AsS.
Arsenopyrite	Dk. gray with rough surface remains	Irid. dk. bn. remains	—	—	—	Ft. bn. rubs clean	—	—	—	—
Chloanthite	Dk. gray remains	Lt. dk. bn. remains	—	—	—	Dk. gry remains	Dk. gry remains	Lt. bn. remains	Dk. bn. remains	Ft. bn. or neg.
Cobaltite	—	—	—	—	—	—	—	—	—	—
Gersdorffite	Dk. gray rough surf. remains	Bn. gry. remains	—	Lt. dk. bn. remains	—	Bn. irid. rubs lighter	—	—	Lt. bn. remains	—
Loellingite	Gry. diff. remains	Gry. diff. remains	—	—	Ft. bn. diff. remains or neg.	Lt. Dk. bn. diff. rubs lt. bn. or neg.	Ft. bn. diff. remains or neg.	—	Lt. dk. bn. diff. remains	—
Rammelsbergite	Dk. gry. blk. remains	Dk. gry. blk. remains	Lt. bn. remains	—	Dk. bn. remains	Lt. dk. bn. remains	Lt. dk. bn. remains	Lt. bn. remains	Lt. bn. stays	—
Safflorite	Dk. gry. remains	Dk. gry. remains	Ft. bn. remains	—	—	—	—	—	—	Lt. bn. diff. remains
Skutterudite	Lt. dk. gry. remains	—	—	—	—	—	—	—	—	—
Smaltite	Dk. gry. remains	Lt. bn. remains	—	—	—	Lt. gry. remains	Lt. bn. remains	—	—	Ft. bn. or neg.

It will be noted that the nickel minerals form a solid block at the top of this rough scale, with the iron and cobalt minerals occupying the intermediate and lower positions. Since all of the nine minerals of this group, with the exception of cobalitite and skutterudite, are etched by dilute nitric acid, this latter reagent forms a good medium of differentiation for these two minerals. In addition, as already outlined in a previous paper by the author,[12] concentrated nitric acid will attack the skutterudite and leave the cobaltite practically unaffected. In this way both these minerals may be differentiated from the other seven of the group and may be distinguished the one from the other.

To present a summary of the results obtained by the various reagents which were found to be successful in separating some of the minerals of this group from the remainder, Table I. has been prepared.

In Table I. very nearly the same order of susceptibility is noted as before, with the first mineral and the last four in the same order but with the remaining four minerals, chloanthite, loellingite, gersdorffite, and smaltite, changing their relative positions slightly. Of the list of ten reagents enumerated in this table, it is not suggested that it is necessary to use all of them for diagnostic purposes. The dilute and conc. nitric acid solutions of the first two columns would seem to be almost indispensable, while the addition of the potassium permanganate, saturated ferric chloride, and aqua regia plus FeAsS, and aqua regia plus (Co, Ni) AsS solutions, should make it comparatively easy to determine any mineral of this group. The remaining four solutions might be used as extra confirmation or checks on the determinations carried out by means of the first-named six. The following table, which includes only these six reagents, demonstrates their differentiating power as applied to the minerals of this group.

[12] Thomson, E., Univ. of Toronto Studies, Geol. Series, No. 20, 1925, pp. 48–65.

	Conc. HNO_3.	Dil. HNO_3.	Pot. Perm.	$FeCl_3$ (Sat.).	A.R. FeAsS.	A.R. (Co, Ni)AsS.
Arsenopyrite	+	+	−	−	−	−
Chloanthite	+	+	−	−	+	+
Cobaltite	−	−	−	−	−	−
Gersdorffite	+	+	+	−	−	−
Loellingite	+	+	−	+	−	−
Rammelsbergite	+	+	−	+	+	−
Safflorite	+	+	−	−	−	+
Skutterudite	+	−	−	−	−	−
Smaltite	+	+	−	−	−	+

Since the dependability of these different etching reactions rests on the purity of type minerals used, it may be well at this stage to give descriptions of these type specimens, including analyses of carefully selected material.

Arsenopyrite.—Specimens of this mineral were used from two localities, one from Marmora, Ontario and the other from Hedley, B.C. No quantitative analyses were carried out on either, but careful qualitative tests in each case gave negative results for cobalt and nickel and strong positive reactions for iron, arsenic, and sulphur. It seems reasonable to suppose, therefore, that both specimens consisted chiefly of the mineral arsenopyrite. The specimens from both localities showed identical reactions to the new microchemical reagents.

Chloanthite.—It was found to be impossible to secure pure specimens of this mineral as it occurred always in the form of zoned crystals with smaltite or smaltite and skutterudite. This phenomenon had, of course, been observed before by Vollhardt and Baumhauer,[13] whose experiments on these composite crystals were repeated by Walker [14] on specimens from the Temiskaming Mine in the Cobalt region. As these crystals were used as type material for the author's work, a few quotations from Walker's paper may be here inserted. He states in part:

> On some specimens recently obtained from the Temiskaming Mine, Cobalt, Ontario, small brilliant crystals resembling smaltite were observed imbedded in fragments of soft chloritic or micaceous country rock in-

[13] *Op. cit.*

[14] Walker, T. L., *Amer. Miner.*, vol. 6, pp. 54–56.

cluded in the vein. As is well known, smaltite and chloanthite closely resemble one another and are commonly intergrown in the same crystal. It was with a view to determining the relative purity of the supposed smaltite that this examination was undertaken.

An analysis made on the powdered mineral is shown in I. The molecular ratio for (Co, Ni, Fe) to (As, S) appears to be nearly 2.5, so that from this analysis the composition would be well represented by the formula $(Co, Ni, Fe)_2(As, S)_5$. Remembering the work of Vollhardt with reference to intergrowths of smaltite and skutterudite, an attempt was made to determine by dissolving part of the powder whether it could be demonstrated that the soluble part had a different composition from the less soluble portion. Some of the same powder was treated with hot 50 per cent nitric for three minutes, and water was then added to interrupt the action. About half the powder was dissolved. The insoluble part was weighed and the weight of the dissolved part was obtained by difference. The analysis of the soluble part is shown in II., the composition of the insoluble part (obtained by calculation) in III.

	I			II			III		
	Per Cent.	Mol. Ratio.		Per Cent.	Mol. Ratio.		Per Cent.	Mol. Ratio.	
Co	20.57	.3506	.4149	20.89	.3558	.4504	20.18	.3437	.3784
Ni	1.31	.0223		2.54	.0433		0.11	.0018	
Fe	2.35	.0420		2.87	.0513		1.84	.0329	
S	1.25	.0391	1.0367	0.99	.0309	1.0017	1.50	.0469	1.0667
As	74.72	.9976		72.71	.9708		76.38	1.0198	
	100.20			100.00			100.01		

From the above molecular ratios it may be observed that the soluble part is most basic and the insoluble part most acidic, thus:

Soluble portion (Co, Ni, Fe) : (As, S) : : 1 : 2.2.
Original mixture (Co, Ni, Fe) : (As, S) : : 1 : 2.49.
Insoluble portion (Co, Ni, Fe) : (As, S) : : 1 : 2.82.

In Hintze's Mineralogie only one of the 55 analyses of smaltite-chloanthite contains as high a percentage of cobalt as the material here analyzed. In most of the cases, Hintze's analyses correspond to complex intergrowths of smaltite-chloanthite with a mineral containing a considerable excess of arsenic. In 39 of the 55 analyses the excess of arsenic (and sulphur) beyond the ratio required for a smaltite-chloanthite mixture is so great, that there must also be present some mineral more

acid than those usually assumed. The most reasonable explanation is found in regarding such samples as composed of smaltite-chloanthite and skutterudite. The general distribution of the last-named mineral in what has often been regarded as smaltite-chloanthite can scarcely be doubted.

The results here brought forward confirm those of Vollhardt and Beutell, that skutterudite and smaltite-chloanthite are capable of forming isomorphous intergrowths in all proportions and the theoretically pure individual minerals are almost unknown.

Besides the material from the Temiskaming Mine, composite crystals from the Silver Bar Mine were also used. While these were not analyzed, they show identical microchemical reactions with the others, the same zonal structure, and gave strong microchemical tests for both cobalt and nickel. Less pure material from the same mine was examined and the description is included along with those from other localities.

Cobaltite.—As type material for this mineral, crystals from the Columbus Mine, as described by DeLury[15] were used. These crystals are described by DeLury as follows:

The mineral crystallizes in cubes and octahedra, but usually in combinations of these two forms.

Some of these crystals were carefully cleaned and examined on a Goldschmidt two-circle goniometer.

	I	II	III
Co	29.10	29.17	35.54
Fe	4.55	4.72	—
Ni	0.97	1.68	—
As	44.55	44.77	45.18
S	20.73	20.23	19.28

An analysis was made of powder obtained from carefully cleaned crystals. The result of this analysis is presented in Column I. For purposes of comparison an analysis of cobaltite from Nordmark (Flink) is given in II., while in Column III. the percentage composition corresponding to the formula CoAsS, is appended.

Crystals from the same mine taken from the small collection of them left by Dr. DeLury for the Royal Ontario Museum were

[15] DeLury, J. S., *Amer. Jour. Sci.*, 21, (4) pp.275–6, 1906.

mounted, polished, and examined. After careful qualitative tests had been carried out, revealing the presence of Co, As, and S in large amounts and the presence in very small quantities of Ni and Fe, it was assumed that these crystals corresponded fairly closely to the analyzed material and could safely be used as type specimens. Some of these crystals showed more nickel than others, as was indicated by their zonal character, brought out by etching with dilute nitric acid. Others seemed to contain but little of this metal. In addition crystals from the type locality in Nordmark were also used. These failed to give any qualitative microchemical reaction for nickel, but gave strong positive tests for cobalt, arsenic, and sulphur.

Gersdorffite.—For this mineral a specimen from the type locality at Lobenstein in Thüringen was used. No analysis was made of this material but strong qualitative tests for Ni, As, and S and negative reactions for cobalt and iron, indicated the comparative purity of this material. In addition, analyzed specimens from the Garson Mine, Sudbury, Ontario, were also used as standards. The analysis of this material carried out by W. Gerrie, shown in Column I., the analysis of Lobenstein material from Dana's System of Mineralogy, shown in Column II., and the theoretical percentages as given by the formula NiAsS, in Column III., are given below:

	I.		II.		III.
S	17.95		18.96		19.3
As	47.70		46.12		45.3
Fe	8.02	34.10	1.81	35.45	
Ni	22.35		33.04		35.4
Co	3.73		0.60		
Ins.	0.80		Cu+Sb 0.44		
	100.55		100.97		100.0
	S.G. 6.06		S.G. 5.95		S.G. 5.6–6.2

From these figures it will be noted that the Garson Mine material differs from the other two only in a larger percentage of cobalt and iron replacing the nickel. Variation from the regular etching reactions was noted in but one specimen of this mineral, de-

tailed mention of which will be made later in connection with the description of individual specimens from the mines now operating at Cobalt.

Loellingite.—The original specimen of this mineral from the Keeley Mine, South Lorraine, as described by Bell and Thomson,[16] representing that part of the original material prepared for analysis but left for microscopic examination, was used for type material. A brief description, with analysis included, is appended:

No. 6.—This is from No. 20 vein on the 7th level south. It presents a massive to fibrous radiating appearance in a hand-specimen, the rhombic character being in evidence in most parts. Its lustre is metallic to sub-metallic, while the color on a fresh surface is tin-white, tarnishing readily to a bronze, or rarely bright blue. In polished section this material is found to consist very largely of rhombic crystals of loellingite, with minor amounts of smaltite and cobaltite. The loellingite forms the major part of this section, with the other two minerals present in subordinate amounts. The analysis is as follows:

	Per Cent.	Mol. Ratio.	CoAsS.	$FeAs_2$.	$CoAs_2$.
Fe	22.24	.398	—	.398	—
Co	5.84	.099	.030	—	.069
As	68.84	.918	.030	.796	.094
Sb	0.26	.002			
S	0.96	.030	.030	—	—
	100.36		4.98%	82.00%	11.10%

In addition to this material other specimens from the Trout Lake Mine, South Lorraine, were used. While no analysis was made of this material, it was at least as pure as the Keeley specimens since qualitative tests gave negative results for both cobalt and nickel, and strong reactions for iron and arsenic. From microscopic examination the only associated minerals appeared to be native silver and tetrahedrite, both of which appeared in the form of tiny stringers cutting the loellingite crystals.

Rammelsbergite.—In the case of this mineral a specimen from the Keeley Mine was selected. It represented part of the material

[16] Bell, J. M., and Thomson, E., Univ. of Toronto Studies, Geol. Ser. 17, pp. 31–2, 1924.

used by Walker [17] in his work on the Keeley ores. This selected specimen seemed from microscopic evidence to consist of practically pure rammelsbergite. In order that this might be confirmed by chemical evidence, part of the polished section used was analyzed by T. E. Rothwell with the following results:

	Per Cent.
Cobalt	2.26
Nickel	25.66
Arsenic	66.57
Iron (met.)	0.35
Sulphur	1.82
Silica	3.42
	100.08

If the silica be left out of consideration and the remaining constituents be recalculated on the basis of 100 per cent., the following figures are obtained:

	Per Cent.	Mol. Ratio.	$NiAs_2$.	CoAsS.	Co_2S_4.
Cobalt	2.33	0.040	—	0.024	0.016
Nickel	26.45	0.448	0.448	—	—
Arsenic	68.63	0.912	0.896	0.024	—
Iron	0.36	0.006	—	—	0.006
Sulphur	1.88	0.059	—	0.024	0.035
	99.65		91.5%	2.24%	6.26%

This confirms the microscopic evidence that this specimen consists principally of the mineral rammelsbergite. Neither cobaltite nor linnaeite appear in the polished section as seen under the microscope, but, since they represent only 8½ per cent. of the total this is not at all surprising. It is not suggested that these minerals must be present but rather they are used as convenient explanations of the remaining small percentage.

Safflorite.—A reasonably pure specimen of this mineral proved to be very difficult to find. After trying many polished sections, a specimen was secured, through the kindness of E. W. Todd, representing the original material from Gowganda described and

[17] Walker, T. L., Univ. of Toronto Studies, Geol. Series, No. 20, pp. 52–3, 1925.

analyzed by him in his report on the district.[18] Todd's description of this material is appended herewith:

Specimen No. 5.—A massive arsenide type of low-grade silver vein from the 585 ft. level north of the winze in the Miller Lake O'Brien Mine consists of small spherical masses of tin-white arsenides rather closely grown together, with uniformly-distributed calcite filling intervening spaces. The metallic minerals are safflorite, loellingite, cobaltite, and a small amount of chalcopyrite. The cobaltite shows up plainly on the etched surface, as small nests of minute cubic crystals scattered through the loellingite and safflorite, which forms intimate intergrowths of radiating blades. The close association of the rhombic mineral loellingite with the cobalt mineral in the intergrowths points to the probability of the latter being safflorite rather than smaltite, the cubic representative of the same composition. The analysis and the percentage composition are as follows:

Analysis		Ratios of Metallic Minerals.	
			Per Cent.
Iron	6.69	Safflorite	58.5
Cobalt	13.97	Loellingite	30.9
Nickel	0.53	Cobaltite	8.6
Sulphur	1.71	Chalcopyrite	1.8
Arsenic	50.70	Silver	0.22
Antimony	1.05		
Copper	0.40		
Silver	0.17		
	75.22		100.02

In this and following analyses, which do not approximate a total of 100 per cent., the balance in most cases consists of calcite and a little quartz. Impurities likely to interfere with the calculation of the proportions of arsenides in most instances are negligible, as proved by chemical tests on the gangue minerals and by microscopic examination. The recalculations serve to check the accuracy of the microscopic determinations and show approximately the relative amounts of metallic minerals. In determining the proportions of the minerals in the table above, the antimony and nickel are placed with the arsenic and cobalt respectively as no minerals containing these elements were observed in the section.

In examining Todd's specimen above-mentioned, the writer was able to confirm his impression that the cobalt mineral was orthorhombic and that the other predominant mineral was loellingite.

[18] Todd, E. W., *Ont. Dept. of Mines*, vol. 35, Pt. 3, p. 68.

It is distinctly unusual to find safflorite so free of the nickel mineral, and the presence of so much loellingite did not interfere in any way with the use of this mineral as a reliable standard for safflorite.

Skutterudite.—While this mineral occurs in composite crystals with smaltite-chloanthite, it also appears in simple crystals by itself. Type material was again selected from the collection of Keeley Mine, South Lorraine, specimens of Bell and Thomson,[19] that portion of the material screened for analysis but left for microscopic examination being used. A brief description from their paper is here inserted:

No. 6B.—This is from No. 20 vein on 7th level south. It is highly altered material of a friable consistency, with bright metallic material in a dull siliceous cement. Some parts of the metallics show spots of the bronze or blue tarnish, so characteristic of loellingite, but for the most part the metallic constituents are tin-white in color, and are in the form of tiny crystals of apparent cubic habit. Under the microscope, it is found to consist almost entirely of skutterudite, with minor amounts of loellingite. The section forms an excellent type specimen of skutterudite, as the amount of loellingite observed was very small. The analysis follows:

	Per Cent.	Mol. Ratio.	NiAsS, CoAsS.	$FeAs_2$.	$CoAs_2$.	$CoAs_3$.
Iron	3.56	.064	—	.064	—	—
Cobalt	17.66	.299	.010	—	.005	.284
Nickel	0.66	.011	.011	—	—	—
Bismuth	0.06					
Arsenic	75.70	1.011	.021	.128	.010	.852
Sulphur	0.66	.021	.012	—	—	—
Insol.	1.64					
	90.94		3.44%	13.09%	1.05%	80.57%

Such small percentages of cobaltite, gersdorffite and smaltite might easily be missing in the microscopic section examined and the slight discrepancy between the chemical and microscopic results may be accounted for in this way. The specific gravity was found from two determinations on the pycnometer to be 6.582 and 6.578. The value, as obtained by calculation is 6.693.

[19] *Op. cit.*, pp. 32–33.

Smaltite.—The same specimens were used for this mineral as for chloanthite, since, as already mentioned, it was found impossible to obtain specimens either of pure chloanthite or of pure smaltite.

On account of the prevalence of carbonate gangue in most of the specimens, considerable difficulty was experienced in treating the polished sections with different microchemical reagents. This was overcome by dissolving out the carbonate with dilute hydrochloric acid to a depth of 1–2 mm. and replacing it either with Canada balsam or sealing wax. The wax was found to be slightly more effective than the balsam. In order to provide for complete filling of the cracks and cavities in these sections, melted wax was poured over the leached surface and the treated specimen was then placed in an oven and allowed to remain for 15 to 20 minutes, at a temperature of about 100° C. The specimen was then repolished, giving a surface of metallic minerals, sealing-wax, and such rock-forming minerals as quartz that resisted the reaction of the leaching agent. The etching reactions on the metallic minerals could then be noted without the weakening of the reagent caused by chemical action on the susceptible carbonate.

Having obtained suitable differentiating reagents for these minerals, the next part of the research deals with the determination of their quantitative proportions in the ores from Cobalt, Ont. Before discussing this part of the work, however, a brief summary of the geology of this well-known district will be given, so that the broad petrographic and genetic relationships may be kept in mind.

GEOLOGY.

The silver-bearing rocks of this region may be placed in three main classes, which are given in the usual order of younger to older:

Keeweenawan (Dikes or sills)—quartz diabase.
Cobalt series—metamorphosed sediments—quartzites, slates, greywackes, greywacke congl.
Keewatin complex—volcanic flows of general basic composition.

The Keewatin rocks of this area are, for the most part, basic volcanics corresponding to altered andesites or basalts in composition, but more acid types approaching closely to the felsites are to be found in occasional outcrops. These rocks are in general heavily schisted. In addition to these volcanic types, the Keewatin is also represented in this field by banded iron formations of the usual siliceous type, sometimes with pyrite replacing the common oxide or hydroxides of iron.

The Cobalt series is a sedimentary series of rocks resting unconformably on the volcanics of the Keewatin and consists for the most part of greywackes or greywacke conglomerate with quartzites and slates as minor members of the series. These rocks also have been subjected to considerable pressure and are frequently very schistose in structure. This feature is, of course, especially pronounced in the slate member of the series.

The Keweenawan dikes or sills intrude both the Keewatin volcanics and the sediments of the Cobalt series. The rock of this formation is a characteristic quartz diabase, considerably altered from its original composition. In the Cobalt area it occurs in the form of a large undulating sill, with fairly flat dips. In addition to these rocks a few small aplite dikes appear in some outlying parts of the field. These would seem to represent the complementary acid member to the diabase itself.

All of these rocks, but more particularly those of the Keewatin complex and the Cobalt series, have been shattered and the resultant cracks and fissures have been filled with material deposited by the later ore-solutions. These solutions have probably originated from the same magma as the diabase but are the result of a later phase of its activity. That this movement took place after the diabase had solidified is demonstrated by the occurrence of small veinlets, carrying native silver as well as cobalt and nickel minerals, within the diabase itself.

ORIGIN AND DESCRIPTION OF SPECIMENS USED IN INVESTIGATION.

In the course of the quantitative microscopic determination of the minerals in the Cobalt ores it was hoped that a more intimate

connection might be established between the diabase and the common arsenides and sulpharsenides present in the ore. With this in view, considerable attention was paid to the determination of these minerals and their relative abundance. A representative suite of specimens was then collected from each of the three principal mines now operating at Cobalt. These, as far as possible, were taken from all the accessible levels, and all possible data as to the distances from the nearest point of the diabase sill were obtained. It is to be regretted that this work was undertaken after the camp had started to decline, as it was found impossible to reach many parts of the mine workings. In spite of this handicap, however, the suites of specimens procured, from the workings of the Mining Corporation, the Nipissing, and the O'Brien Mines, constituted a fairly representative cross-section of the ores of this camp. These accurately located specimens were supplemented by others from more generalized localities. Finally to complete the description, the work of other investigators in this field is also recorded, particularly as it relates to the rarer minerals which have been found in but few places.

The three suites of minerals from the chief mines now operating at Cobalt were collected particularly with a view to establishing a possible relation between the metallic mineralization and the proximity to the diabase sill. They were all accurately located in the various mine workings and included twenty-four specimens from the O'Brien Mine and seventeen each from the Mining Corporation and Nipissing Mine workings. In several cases more than one section was made and examined from one locality, where local variation in the mineral composition made this necessary. These sections were determined partly with the aid of the new microchemical reagents previously mentioned and partly by means of physical tests, confirmation as to the metal content of each mineral being obtained from the familiar ferro-cyanide, dimethylglyoxime, and alpha-nitroso-beta-naphthol tests for iron, nickel, and cobalt, respectively. Microchemical tests for copper, lead and antimony, were also used, where less common minerals were encountered.

The accessible workings of the Mining Corporation Mine are from the surface to a depth of 325 ft., those of the O'Brien Mine from the surface to 716 ft., while the Nipissing Mine workings are mostly at or near the surface but extend in a few instances to a depth of 300 ft. From this it will be seen that the two former mines give data for greater depths below the present surface. It

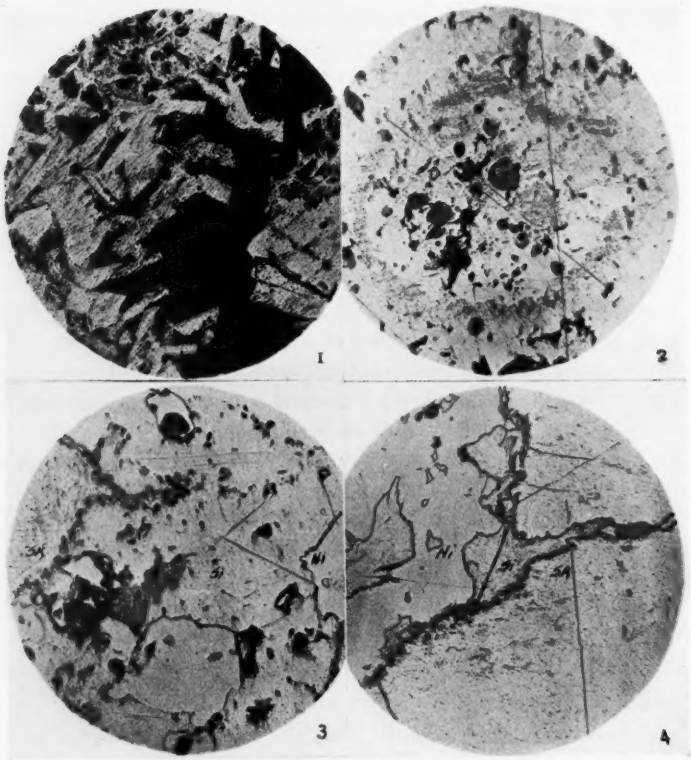

FIG. 1. Spec. M–11. Lt. gray, marcasite; dk. gray, gangue. × 105.
FIG. 2. Spec. N–7. Lt. gray, safflorite; dk. gray, cobaltite; black, gangue. × 45.
FIGS. 3 and 4. Spec. N–12. Si, native silver; Ni, niccolite; Sk, skutterudite; A, argentite; black, gangue. × 105.

MINING CORPORATION SUITE.

No.*	Location.	Major Met. Minerals.	Minor Met. Minerals.
M-1.....	Cobalt Lake below 3d level (210′), 10′ below contact with slate.	Safflorite, cobaltite, silver	Loellingite, skutterudite, argentite, arsenopyrite
M-2-a.....	Cobalt Lake. Vein 4, 3d level (200′), at contact, slate above, Keewatin below.	Loellingite, rammelsbergite, safflorite, skutterudite, smaltite, chloanthite	Tetrahedrite, cobaltite, arsenopyrite
M-2-b.....	Cobalt Lake. Vein 4, 3d level (200′), at contact, slate above, Keewatin below.	Chalcopyrite	Arsenopyrite, tetrahedrite, safflorite, rammelsbergite
M-3.....	Vein 2, east end, 4th level (250′), Cobalt Lake.	Cobaltite, safflorite, skutterudite	Niccolite, loellingite, tetrahedrite, chalcopyrite, pyrite
M-4.....	Fault vein, south-west end, 4th level (250′), at reverse fault.	Cobaltite, rammelsbergite	Safflorite, arsenopyrite, loellingite, smaltite, chloanthite
M-5.....	Vein in cross-cut from vein 2, east end, to fault vein, Cobalt L., 4th level (250′).	Niccolite, breithauptite	Cobaltite
M-6.....	Fault vein, 5th level (325′), foot-wall side 50′ west of ore chute.	Cobaltite, skutterudite, safflorite, tetrahedrite	Rammelsbergite, loellingite, chalcopyrite
M-7.....	Vein 34, Townsite (137′).	Arsenopyrite, cobaltite	Gersdorffite, chalcopyrite, tetrahedrite
M-8.....	Vein 15, Cobalt Lake, 2d level (110′).	Safflorite, niccolite, silver	Arsenopyrite, skutterudite, argentite, chalcopyrite, cobaltite
M-9.....	Vein 11, Cobalt Lake, 2d sub-level (125′).	Silver, niccolite, skutterudite, arsenopyrite, cobaltite	Argentite, chalcopyrite, tetrahedrite
M-10-a....	P Stope, Townsite, 2d level (125′).	Silver, cobaltite, safflorite	Arsenopyrite, rammelsbergite, niccolite
M-10-b....	P Stope, Townsite, 2d level (125′).	Cobaltite, rammelsbergite, safflorite, silver	Skutterudite, smaltite, chloanthite, niccolite
M-11-b....	Vein 1, X fault, Cobalt Lake, 2d level (125′).	Marcasite	

* The numbers are arranged with the lower digits corresponding to the upper levels and the higher to the lower levels.

In a vug about 3″ × 3″ and 1½″ deep the mineral marcasite, which has not heretofore been described for the district, was found with the familiar radiating to sub-radiating structure (Fig. 1). The polarizing microscope showed its anisotropic character very distinctly. This appears to be of much later deposition than most of the other metallic minerals found in this silver camp.

must be borne in mind, however, that the significance of this may not be great since the controlling factor was not the depth below the present surface but rather the depth below the diabase sill. The latter depth, for each of the mines, is estimated to be 75–390 ft. for the Mining Corporation, 0–300 ft. for the O'Brien, and 200–1,100 ft. for the Nipissing. From this it will be seen that the specimens from the Nipissing Mine furnish data in the range from 400–1,100 ft., for which range there are no specimens from the Mining Corporation or O'Brien mines.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
M-12.....	Vein 1, X fault, 200′ east of No. 11 (140′).	Loellingite	Cobaltite, chalcopyrite
M-13.....	Cobalt Lake, vein 25 (75′).	Silver, niccolite, cobaltite, skutterudite	Arsenopyrite, argentite
M-14.....	Vein 34, Townsite (137′).	Pyrite	Chalcopyrite
M-15.....	Vein 1, Cobalt Lake, 2d level (125′).	Silver, niccolite, skutterudite	Cobaltite, tetrahedrite, breithauptite, chalcopyrite
M-16.....	Vein 4, Cobalt Lake, below 3d level.	Silver, skutterudite, cobaltite	Pyrite, smaltite, chloanthite, loellingite
M-17.....	Townsite, 2d level just above Keewatin Iron Formation (100′).	Arsenopyrite	Cobaltite

NIPISSING MINE SUITE.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
N-1.....	Small open pit. On surface.	Rammelsbergite, skutterudite, gersdorffite, cobaltite	Safflorite, silver, smaltite, chloanthite, argentite, marcasite
N-2.....	No. 25 open cut, west end, 30′.	Safflorite, skutterudite	Rammelsbergite, loellingite, smaltite, chloanthite
N-3.....	Vein 54, east end, open cut (50′).	Native silver	Argentite
N-5.....	19 shaft, 19/10 drift, 50′.	Cobaltite, arsenopyrite	Silver, hematite
N-6.....	No. 49 open cut, west end (40′), close to Keewatin contact.	Safflorite	Rammelsbergite, tetrahedrite, loellingite cobaltite
N-7-a.....	Shaft 19, drift 19/23 (100′).	Niccolite, cobaltite, safflorite	Gersdorffite, rammelsbergite, silver

The three major constituents are intimately associated in this section frequently in the familiar concentric arrangement with cobaltite usually occupying the outer portions and either niccolite or safflorite or both at the centers (Fig. 2).

FIG. 5.

FIG. 6.

FIG. 7.

FIG. 8.

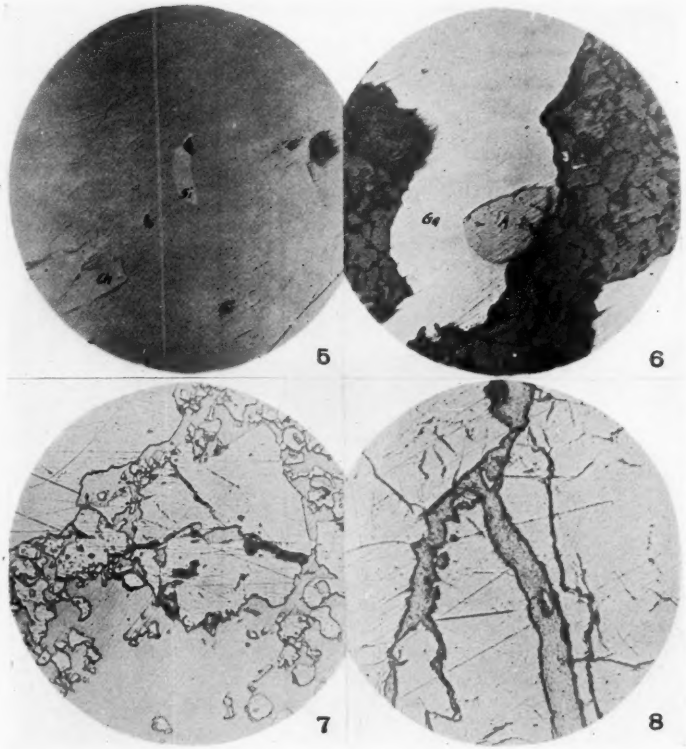

FIG. 5. Spec. O–11–A. Ga, galena; Si, native silver; Ch, chalcopyrite; black, gangue. × 105.

FIG. 6. Spec. O–11–A. Ga, galena; A, argentite; dk. gray, gangue. × 105.

FIG. 7. Spec. O–15–A. Lt. gray, arsenopyrite; dk. gray, chalcopyrite; black, gangue. × 105.

FIG. 8. Spec. O–11–B. Dk. gray, galena; lt. gray, safflorite; black, gangue. × 105.

These three mines, being located at the three points of a triangle, with O'Brien at the north-east, Nipissing at the south-east, and Mining Corporation at the north-west, corners, furnish a fairly good cross-section of the ore of the immediate Cobalt district. The minerals in these sections will now be listed, with emphasis placed on the rough quantitative proportions of the different mineral constituents, at least to the extent of differentiating the major from the minor constituents.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
N-7-b.....	Shaft 19, drift 19/23 (100').	Cobaltite, gersdorffite, safflorite, rammelsbergite	Arsenopyrite
N-8.....	No. 49 open cut, n.e. branch vein, east end.	Silver	Loellingite, niccolite, cobaltite
N-9.....	92 vein, surface, west end, open cut.	Silver, cobaltite, rammelsbergite	Safflorite, skutterudite, smaltite, chloanthite, tetrahedrite, marcasite
N-10.....	Side vein, from 92 vein surface.	Arsenopyrite	Cobaltite, gersdorffite
N-12.....	63 Shaft, 108 vein, west end, top of stope (100').	Silver, niccolite, skutterudite, breithauptite	Cobaltite, argentite

The native silver, niccolite, and breithauptite are intimately associated in this section in irregular veins or blebs (Fig. 4) while the skutterudite fills in the spaces between the areas of these minerals. A small amount of cobaltite is associated with the skutterudite, and a fair amount of argentite is associated with the native silver (Fig. 3).

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
N-15.....	63 shaft, Kendall vein, east or shaft (30').	Silver, rammelsbergite, safflorite	Cobaltite, skutterudite, smaltite, chloanthite, marcasite
N-16.....	80 shaft, Stope 80/107A (65').	Safflorite, skutterudite	Rammelsbergite, cobaltite, silver, chalcopyrite, tetrahedrite
N-17-a.....	Shaft 73, stope 262 (135').	Chalcopyrite	Arsenopyrite, cobaltite
N-17-b....	Shaft 73, stope 262 (135').		Argentite, niccolite, chalcopyrite, arsenopyrite
N-18.....		Gersdorffite	Galena, skutterudite

Gersdorffite here is the predominant metallic constituent, making up over 80 per cent. of the total. One or two crystals of skutterudite are to be seen, while galena appears as irregular stringers cutting the other metallic minerals. A complete chemical analysis of similar material by W. F. Green and T. E. Rothwell, gave the following results:

	Per Cent.
SiO_2	5.10
Al_2O_3	1.11
CaO	5.90
MgO	trace
Fe_2O_3	5.30
As	41.15
Sb	0.31
S	12.36
Ni	22.01
Co	1.89
Total	95.13

Since some calcite was identified in this analysed material and since the amount of lime available would combine with about 5 per cent. of CO_2, it seems probable that the balance of the 100 per cent. is made up of this gaseous material. The silica and alumina may be attributed to quartz and some ferromagnesian mineral respectively, quartz particularly being a not uncommon constituent of these ores in small quantities. If the proportions of the remaining constituents be figured afresh on the basis of 100 per cent the following results are obtained:

Fe_2O_3	6.39
As	49.58
Sb	0.37
S	14.77
Ni	26.52
Co	2.28
	99.92

From a calculation of molecular ratios this gives approximately 88 per cent. NiAsS, 8 per cent. $CoAs_3$ and a small balance of Fe, As, S and Sb remaining. What combinations of iron, arsenic, and sulphur are represented by this small residue of these elements must remain entirely conjectural. There is a possibility that some at least of the iron is combined

with the alumina to form a ferromagnesian mineral. In any case, the amount of such mineral combinations, if they do exist, represents but 4 per cent. of the total. It seems probable from the quantitative relations that the antimony should be placed with the cobalt to make a slightly antimonial skutterudite. No lead was obtained in the analysis but, since galena was identified in but one of the two specimens examined and not in the analysed specimen, this is not surprising. It occurred in any case in very small quantities. It seems reasonable, therefore, to conclude that this section consists largely of gersdorffite, with small amounts of skutterudite, galena and other constituents undetermined. This section did not react normally to the new reagents, potassium permanganate, and aqua regia with iron, giving negative reactions with both. This represents the one anomaly encountered by the writer in dealing with these reagents. Whether this peculiar behavior is due to the presence of a small amount of antimony, or to the occurrence of galena, or to other unknown constituents present in small amounts, it was impossible to determine. It is possible that the antimony really replaces some of the arsenic in the gersdorffite and the presence of a little of the ullmanite molecule may have this modifying effect.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
N-19-a.....	73 shaft, 73 vein, 3d level (250′).	Skutterudite, smaltite, chloanthite, arsenopyrite	Safflorite, cobaltite, chalcopyrite
N-19-b....	73 shaft, 73 vein, 3d level (250′).	Skutterudite, smaltite, chloanthite, arsenopyrite	Safflorite, chalcopyrite
N-20-a.....	64 vein at 73/4143 cross-cut, level 3 (300′), fault vein.	Safflorite, loellingite, niccolite	Rammelsbergite, skutterudite, smaltite, chloanthite, cobaltite, tetrahedrite, chalcopyrite, bornite, covellite
N-20-b....	64 vein at 73/4143 cross-cut, level 3 (300′), fault vein.	Safflorite, loellingite niccolite	Rammelsbergite, skutterudite, smaltite, chloanthite, tetrahedrite, chalcopyrite, bornite, silver
N-21-a.....	73 vein, 4th intermed. level (300′), 73/498 cross-cut small vein.	Galena, pyrite	

O'BRIEN MINE SUITE.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
O-1.....	No. 1 vein between surface and 100′.	Rammelsbergite, cobaltite, skutterudite	Safflorite, loellingite, arsenopyrite
O-2.....	No. 6 vein (300′), 200′ west of No. 6 winze.	Arsenopyrite, cobaltite	
O-3-a.....	No. 16 vein (360′).	Cobaltite, smaltite	Arsenopyrite
O-3-b.....	No. 16 vein (360′).	Galena	Arsenopyrite, tetrahedrite
O-4.....	No. 6 vein (360′), bottom of winze.	Rammelsbergite, cobaltite	Safflorite, loellingite, native silver, arsenopyrite
O-5.....	No. 6 vein (360′), west stope.	Rammelsbergite, arsenopyrite	Cobaltite, tetrahedrite, chalcopyrite
O-6.....	No. 6 vein (360′), sub-drift west.	Loellingite	Silver
O-7.....	No. 6 vein (420′), 200′ west of junction with No. 19 vein.	Galena	Chalcopyrite, tetrahedrite, silver, chalcocite
O-8.....	No. 19 vein (420′), west drift at fault.	Arsenopyrite	Cobaltite, loellingite
O-9.....	No. 6 vein (420′), end of west drift from stope.	Pyrite, pyrrhotite	Chalcopyrite
O-10.....	No. 19 vein (420′), near junction with No. 6 vein.	Arsenopyrite, cobaltite	Smaltite, chloanthite
O-11-a.....	No. 6 vein at junction with No. 19 vein (420′).	Galena	Chalcopyrite, arsenopyrite, argentite, pyrite, sphalerite, silver, cobaltite

Occasional inclusions of native silver or argentite are to be found in the galena (Figs. 5 and 6).

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
O-11-b.....	No. 6 vein at junction with No. 19 vein (420′).	Arsenopyrite	Safflorite, cobaltite, galena

The metallic minerals are roughly interbanded with the carbonate gangue in this section. The other three metallic minerals are also intersected by veins of galena (Fig. 8) and gangue material.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
O-12-a.....	No. 16 vein (470′), 680 stope.	Arsenopyrite, safflorite, cobaltite	Rammelsbergite, silver
O-12-b.....	No. 16 vein (470′), 680 stope.	Pyrrhotite	Chalcopyrite, argentite
O-12-c.....	No. 16 vein (470′), 680 stope.	Galena	Sphalerite, pyrite
O-13-a.....	No. 6 vein (420′), west drift off stope.	Safflorite, silver	Argentite, arsenopyrite[2] loellingite
O-13-b.....	No. 6 vein (420′), west drift off stope.	Galena, arsenopyrite	
O-14.....	No. 16 vein (680′), east drift.	Chalcopyrite, arsenopyrite	Galena, pyrrhotite, safflorite, cobaltite, loellingite, sphalerite
O-15-a.....	No. 16 vein (680′), east drift.	Chalcopyrite, pyrite, arsenopyrite	Sphalerite, galena, silver

Pyrite and arsenopyrite occur in this section in large crystals, with fairly regular outlines in some cases, while the chalcopyrite fills in the spaces between these two minerals (Fig. 7), having replaced them in part at a later period of deposition. Sphalerite is fairly abundant in one part of the section, closely associated with chalcopyrite, but is practically absent in other parts. It occurs in much the same form as the chalcopyrite and frequently carries small inclusions of this copper mineral and of native silver in it (Fig. 9). Galena and native silver are present in smaller amounts as irregular veinlets or blebs, usually located at the grain boundaries of the other minerals (Fig. 10). These metallic minerals are associated with a carbonate gangue.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
O-15-b.....	No. 16 vein (680′), east drift.	Arsenopyrite, silver, cobaltite	Skutterudite, safflorite, galena, chalcopyrite
O-16.....	No. 16 vein (680′), east stope.	Silver, cobaltite, safflorite	Marcasite, argentite, skutterudite, loellingite
O-17.....	No. 6 vein (480′), end of west drift.	Pyrrhotite, galena	Sphalerite, chalcopyrite

The sulphides in this section occupy small fissures in the country rock or similar openings in a quartz vein. Three sections were examined of this material, but, as the metallic mineralization was the same for each, the results were summarized in one description. Sphalerite is present in the smallest quantity, while chalcopyrite is abundant in one and quite rare in the other two sections, where it occurs only as tiny inclusions following the cleavage directions of the sphalerite (Fig. 11).

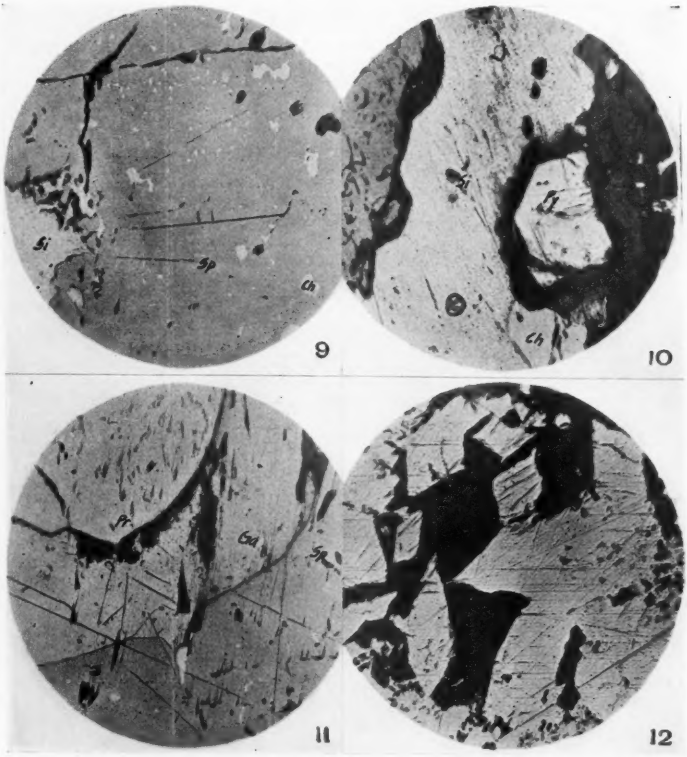

FIG. 9. Spec. O–15–A. Si, native silver; Ch, chalcopyrite; Sp, sphalerite; black, gangue. × 105.

FIG. 10. Spec. M–17. Py, pyrite; Ch, chalcopyrite; Si, native silver; dk. gray, gangue. × 105.

FIG. 11. Spec. O–17. Pr, pyrrhotite; Ga, galena; Sp, sphalerite; white, chalcopyrite; black, gangue. × 105.

FIG. 12. Spec. M–17. Lt. gray, arsenopyrite; dk. gray, gangue. × 105.

No.	Location.	Major Met. Minerals.	Minor Met. Minerals.
O-18-a.....	No. 16 vein (680′), east stope.	Arsenopyrite, safflorite, skutterudite, cobaltite, loellingite	Galena, sphalerite, pyrrhotite, chalcopyrite silver, argentite
O-18-b.....	No. 16 vein (680′), east stope.	Arsenopyrite, cobaltite	Safflorite, silver, argentite
O-19.....	No. 16 vein (680′), end of east drift.	Safflorite, silver	Loellingite, chalcocite, bornite, rammelsbergite cobaltite
O-20.....	No. 16 vein (716′), east drift, contact with diabase 300′ east.	Pyrrhotite	Sphalerite, chalcopyrite, galena
O-21.....	No. 16 vein (770′), west drift.	Galena	Pyrrhotite, chalcopyrite, sphalerite
O-22.....	No. 16 vein (770′), east drift.	Pyrrhotite	Sphalerite, chalcopyrite
O-23.....	No. 16 vein (716′), east drift branch vein contact 250′ to the east.	Pyrrhotite	Galena, sphalerite, arsenopyrite, chalcopyrite, cobaltite

GENERAL CONCLUSIONS FROM DETAILED DESCRIPTIONS.

From a detailed consideration of these seventy-one sections several conclusions are obvious. The first of these is the fact that the old mineralogical title of smaltite-chloanthite for the hard white mineral associates of the native silver and niccolite of this silver region, is no longer applicable, except in a very few instances. As will be seen from a table of quantitative proportions for the hard white metallic minerals of the Cobalt camp, given below in terms of occurrences, the two minerals smaltite and chloanthite are only exceeded in rarity by the nickel mineral

TABLE II.

OCCURRENCES.

	Min. Corp.	Nipissing.	O'Brien.	Total.
1. Cobaltite.....	13½	10½	13½	37½
2. Arsenopyrite.....	6½	5	16	27½
3. Safflorite.....	8½	10	8	26½
4. Skutterudite.....	8½	8½	3	20
5. Rammelsbergite.....	5	7	4	16
6. Loellingite.....	4½	3½	5½	13½
7. Smaltite.....	2½	5	1½	9
8. Chloanthite.....	2½	5	½	8
9. Gersdorffite.....	1½	5	0	6½

gersdorffite. This table was prepared from the results obtained by mineragraphic determinations on the above seventy-one sections, giving a unit value to the major constituents, and half that value to the minor constituents.

As will be seen, the minerals in this table have been listed in order of prevalence and smaltite, chloanthite, and gersdorffite are at the bottom of the list. Cobaltite is much the commonest of these minerals, this result being confirmed by examination of material from other mines in the camp, not now operating. It will be noted too that the high position of arsenopyrite in the list is due largely to the common occurrence of this mineral in the workings of the O'Brien Mine. Skutterudite occurs partly in pure crystals and partly as composite crystals with smaltite and chloanthite. Most of the gersdorffite appears in the form of composite crystals with cobaltite, the notable exception being N–18 where it is almost pure.

The second fact of importance arising from these detailed investigations deals with the common structures observed in each of these nine minerals. While some passing mention has been made of these in individual descriptions, it would seem advisable to summarize the characteristics noted for each of these minerals. They will be discussed in alphabetical order.

1. *Arsenopyrite*—usually occurs in fairly large orthorhombic crystals showing oblong or diamond-shaped section (Fig. 12.) These are commonly arranged as a border round the other arsenides and sulpharsenides and adjoin the gangue material on the other side. In other sections they are scattered through the carbonate, associated with small crystals of cobaltite, or occur as occasional crystals in the masses of other metallic minerals.

2. *Chloanthite*—usually occurs with smaltite as composite crystals with the mineral skutterudite, but occasionally the smaltite-chloanthite is found without the triarsenide. It appears in rough cubic crystals of irregular outline.

3. *Cobaltite*—is most commonly found as a border or rim round rosette-like aggregations of the orthorhombic minerals of this group (Fig. 2). It occurs also as inclusions of small cubic

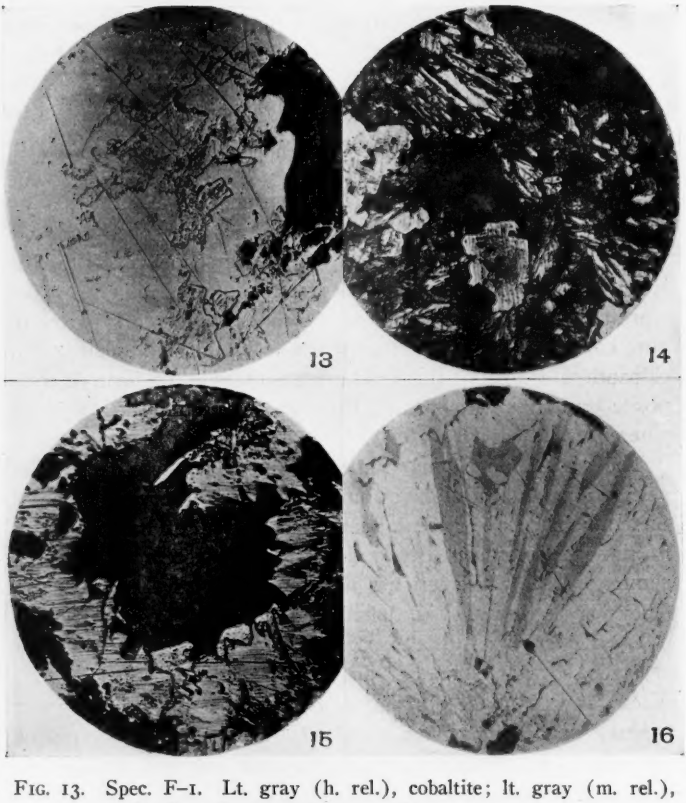

FIG. 13. Spec. F–1. Lt. gray (h. rel.), cobaltite; lt. gray (m. rel.), niccolite; dk. gray, gangue. × 105.

FIG. 14. Spec. N–9. Composite cubic crystals, skutterudite, smaltite, chloanthite; rhombic crystals, rammelsbergite; plain black, gangue; etched with dilute HNO_3. × 50.

FIG. 15. Spec. O–6. White (h. rel.), loellingite; white (l. rel.), silver; dk. gray, gangue. × 105.

FIG. 16. Spec. N–9. Lt. and dk. gray, rammelsbergite; black, gangue; etched with sat. $FeCl_3$. × 105.

crystals in the other metallic minerals (Fig. 13) and in the gangue. Its pinkish white or brownish white color makes it fairly easy of identification. It occasionally forms composite crystals with gersdorffite, when this color becomes more pronounced.

4. *Gersdorffite*—is a rare mineral in this district. It is usually in the form of composite crystals with cobaltite. In N–18, however, where it occurs fairly pure, it is associated with a little later galena and shows, on the weathered surface, a light bluish tarnish. This mineral has been found, from examination of specimens from other localities, to tarnish rather rapidly to a dull gray or black color. Perhaps the difference in tarnish in N–18 may be due to the associated lead ore.

5. *Loellingite*—is found in slender orthorhombic prisms arranged in closely-packed bunches or groups sometimes with a radiating or sub-radiating arrangement (Fig. 15). It is commonly associated with crystals of safflorite or rammelsbergite of similar habit, when it conforms to their rosette-like grouping.

6. *Rammelsbergite*—occurs in slender orthorhombic prisms, associated with safflorite and loellingite, in rosette-like groupings (Fig 16) or with a sub-radiating structure (Fig. 17).

7. *Safflorite*—occurs in much the same forms as rammelsbergite (Fig. 18). The radiating groupings of the safflorite crystals are often elongated and arranged about lines, or really planes, instead of central points. In places it appears in groups of fairly long slender prisms, associated with niccolite (Fig. 19).

8. *Skutterudite*—is found in good cubic crystals, commonly associated with cobaltite. It may also be scattered through the masses of other minerals of this group. In other sections it is found bordering veins or impregnating the adjoining country rock. The mineral appears also as one constituent of composite crystals, the other two being smaltite and chloanthite. In some cases these crystals are zonal in character with alternate zones of triarsenide and diarsenide (Fig. 20).

9. *Smaltite*—occurs in much the same forms as chloanthite described above and is one of the constituents of the zoned composite crystals just mentioned (Fig. 14).

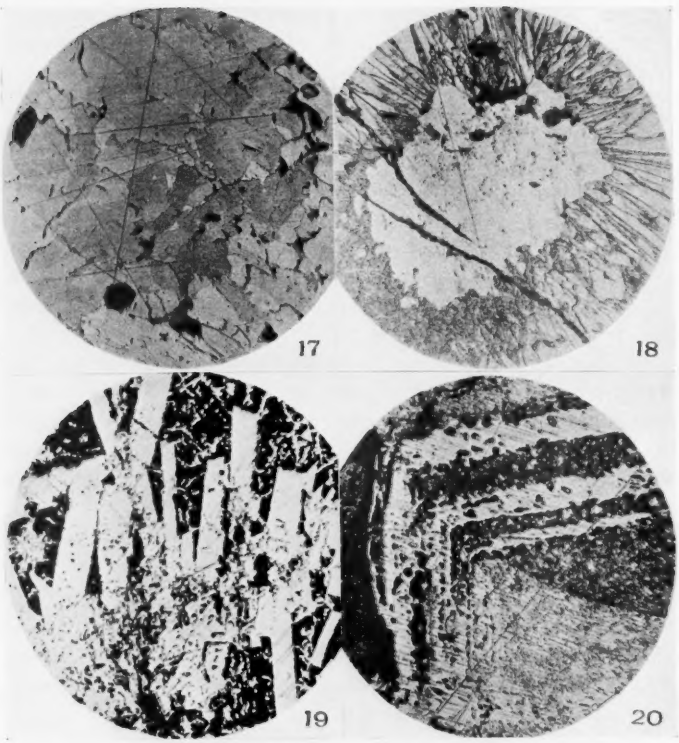

FIG. 17. Spec. N–9. Lt. and dk. gray, rammelsbergite; black, gangue; etched with sat. $FeCl_3$. × 105.

FIG. 18. Spec. M–1. Lt. gray (orth.), safflorite; lt. gray (cub.), cobaltite; black, gangue; etched with dilute HNO_3. × 105.

FIG. 19. Spec. F–1. White prisms, safflorite; black and white mottled, niccolite; etched with $FeCl_3$. × 45.

FIG. 20. Spec. Co–1. Composite crystals, white, skutterudite; dk. gray, smaltite, chloanthite; etched with dilute HNO_3. × 45.

In addition to these nine minerals some of the sections show considerable amounts of the following minerals: argentite, breithauptite, chalcopyrite, galena, marcasite, niccolite, pyrite, pyrrhotite, silver, sphalerite, tetrahedrite, as well as smaller quantities of chalcocite, bornite, and covellite.

The modes of occurrence for niccolite, breithauptite, pyrite, and marcasite are much the same as in other localities. Native silver, dyscrasite, and argentite appear as short, irregular veins or dendritic bodies, whereas the sulphides cut the hard, white, metallic minerals and the gangue material of this district, in the form of small irregular veins or blebs. These metallic minerals are associated chiefly with a calcite gangue, though smaller quantities of a siliceous material are to be found in some specimens. This silica appears to be later, in time of deposition, than most of the carbonate.

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(*To be concluded in next number*).

GENESIS OF THE EMERY DEPOSITS NEAR PEEKSKILL, NEW YORK.

JOSEPH L. GILLSON AND JOSEPH E. A. KANIA.

SUMMARY.

THESE deposits, still mined on a small scale, are located 35 miles north of New York city on the east bank of the Hudson, along and on both sides of the contact of the intruded Manhattan schist (all or partly pre-Cambrian) and Cortlandt (post-Ordovician) intrusives.

Described in 1888 by Williams as contact-metamorphic deposits, they have since been described (G. S. Rogers, 1911) and cited (N. L. Bowen, 1922) as typifying magmatic differentiation by assimilation of the wall rock. The authors present data disproving this assimilation theory and proving that the deposits are contact-metamorphic in origin and were formed by gaseous or liquid emanations from the magma reservoir which passed upwards through the already solid border of the igneous mass, and into the schist.

A detailed mineralogical and petrographical description of the rocks and ores is given.

GENERAL STATEMENT.

The Cortlandt area of post-Ordovician igneous intrusives, dominantly of basic types, contains one of the few emery deposits in America that have been worked commercially. The igneous complex, with its many rock types and interesting border effects, has interested geologists for more than fifty years, and the problem of its contact metamorphism, described by Williams [1] in 1888, was one of the first of such problems to be discussed in American

[1] Williams, G. H., "The Contact Metamorphism Produced in the Adjoining Mica Schist, and Limestones, by the Massive Rocks of the Cortlandt Series, near Peekskill, N. Y." *Amer. Jour. Sci.* (3), vol. 36, pp. 254–269, 1888.

geological journals. Later, however, when the idea of reaction by assimilation was introduced, the conception of contact metamorphism was forgotten, and the emery deposits were selected as a classic example of such reaction and assimilation.

The emery deposits lie mostly southeast of Peekskill, a town on the east bank of the Hudson River, about 35 miles north of New York City, and within the West Point Quadrangle (Fig. 1).

FIG. 1. Outline map of New York State, showing location of Peekskill.

The Cortlandt series occupies a compact area of about twenty square miles, lying within an area of old crystalline rocks, all or partly of pre-Cambrian age. The wall rock almost entirely surrounding the intrusives is the Manhattan schist, a highly foliated quartz-mica schist, named for its occurrence on Manhattan Island, on which lies the city of New York. The emery is a fine-grained aggregate of corundum, spinel, and magnetite in varying proportions and is found only at or near the igneous contact, and both in the metamorphosed schist and in the endomorphosed igneous rock. The ore does not occur in large masses, and in working the pits much hand sorting was necessary.

The igneous rock found associated with, or cropping out near the ore deposits, is in most places a basic norite or pyroxenite

carrying less than 15 per cent. of feldspar. Many narrow sills or stringers cut the schist and inclusions of the schist occur within the igneous rock.

The emery deposits commercially most important are confined largely to the vicinity of a hill lying about three miles east of Peekskill, marked with light shading on Fig. 2. Because of the

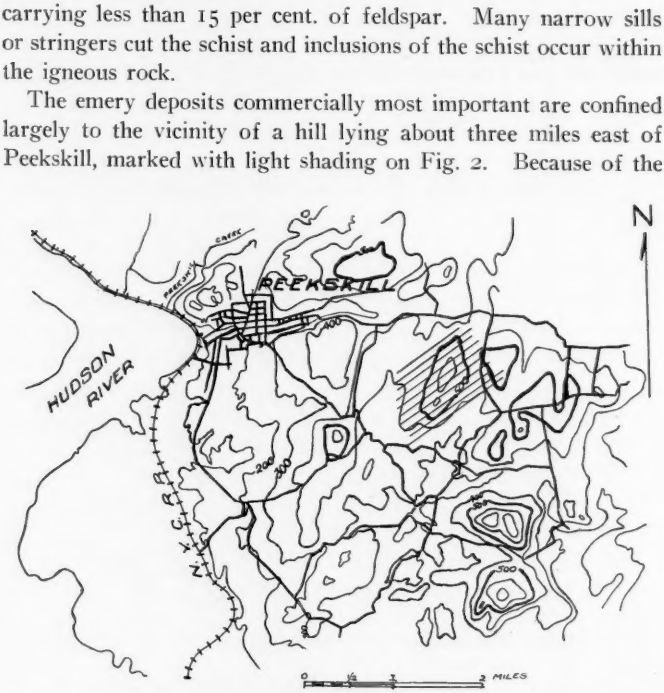

FIG. 2. Topographic map of Peekskill district. Area of principal emery pits shown with shading. Contour interval 100 feet.

good exposures in the various openings, and abundant material favorable for study lying on the dumps, most of the observations were made in this vicinity.

The theory of origin of these deposits advanced by Rogers[2] was that the emery was formed by the absorption of the Manhattan schist in the basic Cortlandt magma, and because of the reaction of the magma on the wall rock. He based his conclusions largely on a series of experiments that had been made by

[2] Rogers, G. S., "On the Geology of the Cortlandt Series and its Emery Deposits," *Annals N. Y. Acad. Sci.*, vol. 21, pp. 11–86, 1911.

Morozewicz.[3] Bowen [4] took these deposits as a typical example of his ideas of the reaction of an aluminous wall rock on a basic, and molten, magma.

The present study has shown that the minerals making up, and associated with the emery, had formed, not when the molten magma was near by, but rather after the consolidation of the marginal facies of the igneous rock. The emery and related minerals are typical contact-metamorphic minerals and all formed in a sequence in the wall rock and already solid igneous rock as the result of the passage of emanations given off during a long period of time from the cooling magma chamber in depth.

GENERAL GEOLOGY.

The following summary of the general geological features is taken from the detailed studies of Rogers [5] and Williams.[6] The Cortlandt series was named by J. D. Dana from Cortlandt township, Westchester county, New York. Similar rocks are found in a small patch covering less than one quarter of a square mile on the west bank of the Hudson River, at Stony Point; another extension is found at Rosetown, New Jersey, and two areas of similar rocks, probably related to the Cortlandt series, occur in western Connecticut.

In the Cortlandt area examples of nearly every group of intrusive igneous rocks exist. The age of the series is determined by the fact that the intrusion took place after the period of intense metamorphism in the Ordovician. Rock types represented are: granite, syenite, sodalite syenite, diorite, gabbro, norite, pyroxenite, peridotite and hornblendite. In addition to the plutonic types, hypabyssal examples occur: aplite, pegmatite, and dacite porphyry.

[3] Morozewicz, J., "Experimentische Untersuchungen über Bildung der Mineralien im Magma," Tschermak's *Min. und Petr. Mitt.* XVIII., pp. 1–90 and 105–240, 1898.

[4] Bowen, N. L., "The Behavior of Inclusions in Igneous Magmas," *Jour. Geol.*, vol. 30, pp. 550–560, 1922.

[5] Rogers, G. S., *op. cit.*

[6] Williams, G. H., *op. cit.*

Of the intrusives, a noritic rock is more widely exposed than any other of the group. True norite is, however, subordinate in occurrence. Rogers distinguished biotite norite, biotite-augite norite; quartz norite, augite norite, hornblende norite, biotite-hornblende norite, and olivine-augite norite. The rock Rogers called pyroxenite is important because all the commercially operated emery deposits have been found in the eastern pyroxenite area. In this study this rock with approximately fifteen per cent. of feldspar was called a " mela-norite."

The Manhattan schist, the country rock into which the igneous complex was intruded, is a highly schistose, quartz-mica-chlorite schist, with soda and potash feldspar, and in places important amounts of a carbonate. Bowen [7] makes a strong point of the composition of the schist as being similar to that of an acid igneous rock.

Close to the igneous contacts the schist is found to have been metamorphosed into an apparently schistose, but actually tightly interlocking aggregate of sillimanite, cyanite, quartz, andesine, biotite, cordierite, and amphibole. Because of its mineral composition and pseudomorphic schistosity, it is similar to the contact-metamorphosed Ellsworth schist described by Gillson and Williams.[8]

The emery, which is a heavy, dense, dark-colored rock consisting essentially of spinel, corundum and magnetite, is not found in large masses of solid ore. It occurs in pods, lenses and vein-like masses, both in the hornfels and the endomorphosed igneous rock. These vein-like masses are in places irregular in form, and may have, as in some of the openings on the west base of the hill shown shaded on the map, very bizarre shapes. In the hornfels many narrow stringers of ore one to two cm. across, occur parallel to the pseudomorphic schistosity. The fact that the ore occurs in this manner is not a new observation, and it is so described by Rogers.[9]

[7] Bowen, N. L., *op. cit.*, page 552.

[8] Gillson, J. L., and Williams, R. M., " Contact Metamorphism of the Ellsworth Schist near Blue Hill, Maine," ECON. GEOL., vol. 24, No. 2, pp. 182–194, 1929.

[9] Rogers, G. S., *op. cit.*, p. 73.

THE NORITE.

The Fresh Mela-norite.—The fresh mela-norite, outcropping near, but never in connection with, the ore bodies, varies from place to place. Near Strangs' upper pit, on the west side of the shaded area, the rock is coarse-grained, black in color, and of striking appearance because of large glistening cleavage faces of poikilitic grains of hornblende. Many of these are 2 cm across. Labradorite forms only about 15 per cent. of the rock. Lower down on the west side of the hill the rock is even-grained, with an average grain size of about 1/10th mm. and hypersthene, augite, and subordinate brown hornblende compose 85 to 90 per cent. of the mineral composition. Farther to the east, on top of the hill, the fresh rock exposed is black, even-grained, and consists of 75 to 80 per cent. of pyroxene with subordinate feldspar. Still further to the northeast, on the same hill, dominantly feldspathic norite was found, the rock consisting principally of labradorite and hypersthene.

Thus the typical fresh norite can be described as a melanocratic hypersthene-labradorite rock with subordinate augite, brown hornblende, and biotite. Accessories are rare.

The fresh norite offers a problem in the genesis of poikilitic minerals, which deserves a passing note here. The large glistening faces of hornblende have been described. Embedded in these large grains are much smaller grains of feldspar and pyroxene. An example is shown in Fig. 3, which was drawn from a grain about 4 mm. long, found in the thin section of a specimen from near Strangs' upper pits on the west side of the ridge. The hornblende, which is a deep-brown variety, common to basaltic rocks, could not have crystallized into this rambling shape as phenocrysts, during the early pyrogenetic stage. The suggestion is advanced here that these poikilitic grains are entirely or partly deuteric, and formed at or after the close of the pyrogenetic crystallization, and partly or entirely by replacement.

The magnetite of the rock is also partly deuteric, as is shown by the shape of the grains in Fig. 4. Pyrrhotite and pyrite also were found with similar forms. Such evidences of deuteric

minerals in the norite indicate that the emanations causing the more evident and complete changes at the borders passed also through the body of the rock, and made some changes there.

FIG. 3. A poikilitic grain of brown hornblende, about 4 millimeters long. A grain of this shape, and much larger than the associated minerals, could not have formed as a phenocryst in the early pyrogenetic stage. It must be entirely or partly deuteric. It occurs in the mela-norite that is not affected by the endomorphism, but indicates that that rock also was acted upon by post-consolidation emanations.

FIG. 4. Deuteric magnetite in the fresh mela-norite. Grains of this shape must have formed at a late stage in the crystallization of the rock, and partly or entirely by replacement.

The biotite in the typical norite has a pleochroism from light yellow to a deep reddish brown, quite different from that in the endomorphosed igneous rock described below. Some of it continued its growth into the deuteric stage, as is shown in Fig. 5,

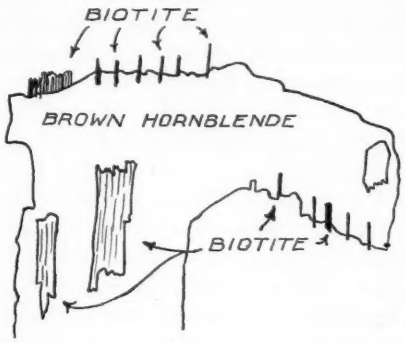

FIG. 5. Needles of biotite formed in the cleavages of the large crystals of brown hornblende in the fresh mela-norite, showing additional action by post-consolidation emanations.

where tiny plates of biotite extend into the cleavage planes of a large hornblende grain.

Apatite, though not an abundant constituent of the mela-norite, was found in small quantities in a few of the thin sections. The grains are relatively large, being from .03 to .5 mms., subhedral in form, and have in most cases a dark color because of abundant microscopic black inclusions. The rarity of the apatite, and its occurrence only in rather large grains, is in marked contrast to the manner of occurrence of apatite in the endomorphosed rock, in which tiny needles are very abundant.

THE ENDOMORPHOSED NORITE.

No ore was found with the fresh hypersthene-labradorite mela-norite just described. It occurs rather with a modified product of that rock that is found only at the margins in contact with the schist. Thus a study of this modified product is most important

in considering the genesis of the emery, but its importance seems to have been previously overlooked, and Rogers noted only in passing that a type, " quartz norite " existed in the district.

The rock has in most cases a fine to medium, and uniform grain. The color of the weathered surface is greenish gray to grayish black, when seen from a distance, but on close examination the rock is best described as having a pepper-and-salt appearance. In some exposures poikilitic feldspar grains, or poikilitic pyroboles occur that are considerably larger than the majority of the other constituent grains. In general, however, the rock is so undistinctive in megascopic appearance that it may be readily understood why its importance was not recognized.

In studying the thin sections, the relation of the endomorphosed product to the fresh melanocratic norite is obvious from the many remnants of the original grains, but important mineralogical changes have occurred. The pyroxene has been more or less completely changed to secondary amphiboles, among which a nearly colorless orthorhombic variety is the most interesting. The calcic labradorite, of which the disseminated black inclusions (so commonly observed in the plagioclase grains of basic rocks) are a characteristic feature, has been partially made over into, surrounded by, cut through, and partially replaced by, a clear glassy feldspar, free from inclusions, that is in most cases andesine, but in a few specimens is oligoclase. The replacement is as clear in some of thin sections of this endomorphosed norite as in those of the Blind Mountain quartz monzonite from Lincoln County, Nevada, described recently by the senior author.[10] A sketch of one such grain is shown in Fig. 6, and veins of oligoclase that cut directly through large labradorite crystals shown in several slides, are certainly also the result of the action of post-consolidation emanations, instead of having crystallized from the original melt by normal zoning.

A considerable number of the feldspar grains appear to have a mottled appearance due to lack of uniform extinction between

[10] Gillson, J. L., " The Petrography of the Pioche District, Lincoln County, Nevada," U. S. Geol. Surv. Prof. Paper 158, pp. 80–82, 1929.

crossed nicols. This was explained by Rogers as being due to crushing. Although in places this is probably true, nevertheless in others the irregular replacement of the grain by a feldspar of nearly the same composition gives the same appearance.

FIG. 6. A grain of plagioclase in the endomorphosed norite. The center is labradorite, but the outside is andesine, and some of the latter had penetrated to the very center of the grain. This fact, and the lopping off of a piece of the labradorite, as shown by the shaded fragment in the upper right-hand corner of the grain indicates that the feature is a replacement and is not normal zoning. Q = quartz.

Quartz occurs in most of the specimens studied, and in many it is very abundant. Proof that some of the quartz replaced pyrogenetic grains is not readily apparent at first glance but is evident upon close observation. The quartz occurs, in places, in veins, and many examples of grains showing other cross-cutting relations were observed. Also, in many of the quartz grains are tiny cavities, in some of which are moving bubbles, probably of carbon dioxide, and in others tiny crystals. These features are known to be of common occurrence in vein quartz, but to the writers' knowledge are absent, or at least rare in pyrogenetic quartz. Furthermore, nearly all the quartz grains contain abundant whisker-shaped grains of rutile, and the presence of this is considered to be a further proof of hydrothermal or pneumatolytic origin of the quartz, as rutile inclusions are commonly found only in quartz of that origin.

The biotite has not the deep red-brown pleochroism of that in the fresh mela-norite, but the deepest tint is a greenish brown of moderate intensity. In places this biotite is abundant, making up a quarter of the mineral composition. In many of the specimens the biotite and secondary amphibole are intergrown.

The secondary amphibole is present in most of the thin sections of the endomorphosed rock, as a replacement of both the augite and the hypersthene. A green uralitic variety occurs in small amounts in a few slides, but the most wide-spread variety is anthophyllite, having the following properties: alpha = 1.652 $\pm$ .003; gamma = 1.672 $\pm$ .003; and colorless.

Graphic (?) intergrowths of spinel in feldspar were noted in places. One such structure is about one mm. long and the cluster, which has an oval shape in section, consists of several score of spinel anhedrons, each roughly .025 mm. in diameter, set in andesine grains that are around .1 mm. long. Except in this cluster, spinel is absent from the slide.

Graphic intergrowths of magnetite in pyroxene were seen in several slides. The magnetite grains have an elongated and curved form in section, and vary in length from .01 to .05 mm. Similar intergrowths have been observed by the senior author in the gabbros and contact rocks of the Adirondack region and deserve more study and thought. It is improbable that they are pyrogenetic structures.

In addition to the occurrence of magnetite just mentioned, it is widely disseminated in the endomorphosed rock. The grains have the common form of deuteric magnetite, and widely disseminated grains of apatite and zircon, and grains of carbonate, pyrrhotite, chalcopyrite, and pyrite, and rare grains of allanite, further testify to the fact that the rock has been affected by emanations rich in the rarer constituents.

It has been stated above, and will be further described below, that emery ore occurs in this endomorphosed rock in veins, pods and irregular masses. (Fig. 7). A few scattered grains of corundum were found in a few of the slides of the modified igneous rock away from the ore, but never abundantly. In other

words, where the emery occurs in the igneous rock, it is in concentrated form, and shows sharp cross-cutting relations with the other minerals.

THE HORNFELS.

The recrystallized Manhattan schist found at the igneous contact was recognized as early as 1888 as a contact-metamorphic product. Bowen, however, described the mineralogical changes as being due to a reaction between the solid sediment and liquid magma, although how the recrystallization could have been affected tens of feet from the contacts was not explained by him. He believed that the old quartz-mica-chlorite schist was turned into a sillimanite rock by the solution from it of those oxides readily soluble in the magma, and what remained was "surplus material" that recrystallized as a sillimanite rock.

Certainly the complexity alone of the mineralogy of these contact zones speaks strongly against such an interpretation.

The hornfels is dark gray to black in places, but where the sillimanite is abundant it has a lighter color, and weathers to a light gray. In many of the emery pits a dark hue with a reddish cast is seen on the hornfels, which stands in marked contrast to the almost black color of the emery ore.

Much of the hornfels is so dense in texture that individual grains cannot be distinguished megascopically, but in many exposures grains of sillimanite, cyanite, garnet, quartz, cordierite, or chlorite, from one to two millimeters in diameter, occur and in a few places much larger crystals of garnet, tourmaline, and cyanite were found.

To one familiar with many examples of contact-metamorphic zones around igneous contacts, the nature and mineralogy of the recrystallized Manhattan schist appear most typical of such zones and, in fact, no other interpretation is tenable. The minerals cordierite, garnet, tourmaline, andesine, sillimanite, staurolite, amphibole, muscovite, biotite, chlorite, spinel, corundum, cyanite, magnetite, pyrrhotite and pyrite found in an intruded rock not elsewhere containing those minerals, and for a considerable distance from the contact, could have formed only be-

cause of the stimulus of gaseous or liquid emanations given off by the magma during its crystallization. Furthermore, it would be an unusual occurrence were it not found that these many silicates had formed mostly after the igneous rock was solid at its border.

Thus, as was to be expected, a study of the boundary relations of the minerals indicates a sequence in their formation which is about as follows: andesine, cordierite, amphibole, sillimanite, corundum, biotite, staurolite, later sillimanite, spinel, chlorite, magnetite and sulphides. This sequence or paragenesis indicates stages or periods in the process of contact metamorphism, resulting from progressively changing conditions of composition and temperature, and indicates further that the mineralization was the result of a parade of solutions that were given off through a long period of magmatic crystallization in depth, with progressively lowering temperature. Such features have been described before,[11] and are very different from a quick partial solution by the molten magma and a recrystallization of the "surplus material."

As is characteristic of all contact-metamorphic zones, a great variety exists among the specimens collected, and a general description must be a composite picture of many diverse rocks. The general mineralogy has been given. Near emery veins, högbomite occurs as an apparent alteration product of the spinel, and in some examples sapphirine is an abundant mineral and is probably the blue pleochroic corundum described by Rogers, for all the corundum seen by the writer is colorless.

The "pseudomorphic schistosity" referred to above is expressed in a banding of the rock, seen not only in the outcrop but apparent also in the thin sections, and is a result of diversity of mineralogy caused by primary differences in composition in the

[11] Gillson, J. L., "Contact Metamorphism of the Rocks in the Pend Oreille District, Northern Idaho," U. S. Geol. Surv. Prof. Paper 158–F, 1929. Spurr, J. E., Garry, G. H., and Fenner, C. N., "A Contact Metamorphic Ore Deposit; the Dolores Mine at Matehuala, San Luis Potosi, Mexico," Econ. Geol., vol. 7, pp. 471–474, 1912. Umpleby, J. B., "Geology and Ore Deposits of the Mackay Region, Idaho," U. S. Geol. Surv. Prof. Paper 97, p. 65, 1917. Eckermann, Harry von, "The Rocks and Contact Minerals of the Mansjö Mountains," *Geol. Fören. Förh.*, 1922, p. 343.

layers of the schist. Although closely banded, and thus appearing schistose, the minerals are in random orientation, and closely intergrown, knitting the rock into a massive whole.

Each of the minerals of the hornfels has certain interesting features worthy of mention. The feldspar, which in most specimens is andesine with a symmetrical extinction at right angles to the 010 zone of 20° to 25°, is not an abundant mineral, and is absent from most of the specimens and dominant in none. It occurs as interlocking anhedra, most of which are under .1 mm. in diameter. Many of the grains appear mottled and, in not a few, a sharp boundary can be seen between a corroded core with a higher refractive index (labradorite), and a rim of andesine. These differences in composition within the grains were perhaps most conspicuous in an emery-bearing piece from one of the pits on the east side of the hill in the shaded area. This piece has many sharply defined emery bands varying in width from ½ to 1 inch, and these cross it with many twists and turns, some of which are the pseudomorphic "drag-folds" of the schist.

Bowen [12] would explain such "feldspathic emery" as coming from "inclusions approaching their final disappearance." Yet the core of the feldspar grains is labradorite, although the plagioclase of the primary Manhattan schist is a sodic variety. Another explanation might be that the feldspathic emery is an intensely endomorphosed norite, but it contains not one of the other minerals of that rock but rather those of the hornfels, and has the "pseudomorphic schistosity" above referred to. The phenomenon seems to be best explained by stating that at an early period, while the magma was still molten and labradorite was crystallizing in it, the solutions passing out into the country rock formed labradorite. Later, with the much more abundant emanations in connection with the endomorphism of the igneous rock, and when its labradorite was being replaced by andesine, then andesine formed in the hornfels.

Cordierite is an expected mineral in an occurrence of this type. It is visible macroscopically in places as bluish, glassy crystals,

[12] Bowen, N. L., *op. cit.*, p. 556.

and in thin section it is made conspicuous by spots of yellow pleochroism, locally of intense color. Most of these occur around tiny zircon grains, sub-microscopic zircon grains, or grains not exposed in the plane of the section, suggesting that the pleochroic halo is due to radio-active decomposition.

The sillimanite occurs in large and small prisms, and is one of the most abundant minerals of the hornfels. Many of the large prisms show an irregularly distributed pleochroism—Z being a dark reddish brown. Smaller laths are colorless. A second generation of sillimanite is recognized because of the replacement of large grains of sillimanite, and of other minerals, by a felt-like mass of small grains. It resembles the fine felt-like aggregates of sericite seen in many examples of hydrothermal wall rock alteration along ore veins.

In no place was corundum seen as an apparent replacement of sillimanite, as demanded by Bowen. So far as could be observed, corundum formed contemporaneously with the larger prisms of sillimanite.

The amphibole is an unusual variety of the orthorhombic group. It is pleochroic in pale tints, a slate gray being the darkest shade. The mineral is optically positive, with an optic angle of about 85° and an axial dispersion of violet greater than rho. The refractive indices are: alpha $= 1.658 \pm .003$, and gamma $= 1.675 \pm .003$. Next to the sillimanite the amphibole is the most abundant mineral of the hornfels.

Staurolite occurs in euhedral and subhedral grains, many of which are found in the chlorite, probably because they formed in biotite, later altered to the chlorite. Cyanite and garnet are not very abundant and deserve no particular mention.

The biotite has a moderately strong pleochroism in greenish brown tints, approximately those shown by the biotite in the endomorphosed igneous rock. The biotite is an abundant mineral of the hornfels, and Rogers mentions the occurrence of many biotite-rich zones close to the emery bodies.

The chlorite is also an abundant mineral, and a very conspicuous one in the emery pits. It properties are as follows:

Optically positive, 2V = about 10°; pleochroism from nearly colorless for Z, to pale green for X; does not show anomalous interference colors; alpha = 1.602, gamma = 1.608.

These properties place it close to the ripidolite of Winchell.[13]

Zircon is wide-spread in the hornfels, and many of the grains are as large as .025 mm. in diameter, although the majority are under .01 mm. Titanite, on the other hand, which should be expected in such an association, was not positively identified in any of the sides. Moreover the absence of the quartz from the hornfels is noteworthy, although some quartz stringers do cut through it.

THE EMERY ORE.

The ore occurs both in the hornfels and in the endomorphosed igneous rock and, so far as the writers observed, as good ore was found in one as in the other. Some of the larger openings are in the endomorphosed igneous rock, others are entirely in the hornfels, the majority expose both types.

Some detailed observations will illustrate the character of the emery and its occurrences.

In Strang's upper pit, the ore is a heavy, dark-colored, fine-grained rock, in which layers or veins of solid emery occur, each of which is about one-quarter inch across. A microscopic study of the gangue identified it as endomorphosed mela-norite, in which the two stages of feldspar formation are conspicuous, and in which secondary amphibole, quartz, zircon, apatite and allanite occur, minerals absent from the fresh igneous rock. The emery consists of corundum, spinel and magnetite. The corundum grains are about 1 mm. in diameter, the spinel grains are smaller. In most cases the spinel grains, rather than those of corundum, adjoin the feldspar. In the emery bands, spinel makes 65 per cent., magnetite 20 per cent., and corundum 15 per cent. The hornfels, a few feet away, contains accessory corundum and magnetite, but no spinel, and contains sillimanite, cordierite, cyanite, staurolite and the chlorite. Eight feet from the first ore zone is another in the hornfels. It consists of grains about

[13] Winchell, A. N., "Elements of Optical Mineralogy," II., p. 380, 1927.

.05 mm. in diameter, and corundum makes up 65 per cent. of the composition, with magnetite 25 to 30 per cent. and högbomite 5 per cent. This mineral is of common occurrence in the ore, as it is in the Virginia emery deposits described by Watson.[14] In the Peekskill ore the properties approximate those listed by Larsen,[15] and although Watson[16] describes the mineral in the Virginia ore as being isotropic, this is apparently a mistake, as Dr. Larsen,[17] who studied Dr. Watson's slides, says that the mineral there had a strong birefringence.

In Strang's No. 3 cut the south wall is endomorphosed melanorite. In contact with it is a typical sillimanite rock in which are bands of emery containing corundum, spinel and högbomite, with magnetite and abundant later chlorite. Lenses and pods of emery of bizarre shape occur in the endomorphosed igneous rock. Fig. 7 illustrates the sharp crosscutting relations of the ore to the igneous rock.

A suite of specimens collected across the igneous contact and the strike of the pseudomorphic schistosity, in a pit in the meadow to the SE of the summit of the hill in the shaded area, illustrates the differences in the mineralogy of the rock types, and in the different bands of the hornfels. The igneous rock is so intensely endomorphosed that no pyroxene remains, all having been replaced by a monoclinic amphibole having a pleochroism from yellow to green to blue. The adjacent hornfels contains disseminated corundum, spinel and magnetite, and many sharply defined bands of nearly pure emery, parallel to the old schistosity. Some of these are wide enough to furnish commercial ore. Besides the emery bands in the hornfels, other bands rich in cordierite and others in sillimanite occur.

Strang's "Horseshoe Pit" on the west base of the hill is also instructive. The hornfels contains well defined bands preserving,

[14] Watson, T. L., "A Contribution to the Geology of the Virginia Emery Deposits," ECON. GEOL., vol. 18, pp. 53–76, 1923.

[15] Larsen, E. S., "Determination of the Non-opaque Minerals," U. S. Geol. Surv. Bull. 679, p. 201, 1922.

[16] *Op. cit.*, p. 69.

[17] Larsen, E. S., private communication.

as a pseudomorph, the old schistosity. Parallel to this are bands of emery some of which are as thin as 1 mm., and such thin layers, continuous for many feet, could not have resulted from magmatic reaction. They do, rather, represent layers of favorable com-

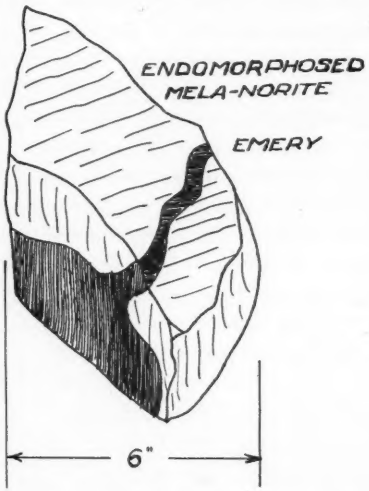

FIG. 7. Sketch of a chunk of ore-bearing norite, lying in the floor of Strangs' No. 3 cut, at the west base of the hill in the shaded area. This shows that the emery occurs in the endomorphosed igneous rock as a sharply cross-cutting vein, not as a remnant of a partially absorbed inclusion of schist.

position along which solutions of progressively changing nature soaked slowly through the rock.

The hornfels contains the orthorhombic amphibole, pleochroic in pale tints, that is so wide-spread, and in addition sapphirine, biotite, sillimanite, cordierite, corundum and magnetite. The sapphirine has the following properties:

Biaxial, negative, 2V moderate; dispersion extreme, $r > \rho$; $C \wedge Z$ about 10°, pleochroism from light green to very deep blue; sections cut nearly at right angles to the optic axis show deep Berlin Blue colors.

The endomorphosed igneous rock makes the inner concave wall of the "horseshoe," and contains lenses and pods of emery, some of which are less than one inch in diameter.

Excellent samples of the occurrence of emery in schist are found in the pits on the summit of the hill. Many sharply defined black emery bands are only ½ to 1 cm. across, and on close examination many places were noted where tongues and protuberances of such narrow bands project laterally into the silicates of the hornfels, and narrow stringers cross from one wider band to a neighboring one several centimeters distant.

Tongues, protuberances and stringers of ore in schist could not have resulted from the reaction of a sillimanite-rich inclusion in the magma some distance away and the crystallization of the "surplus material" as emery. For the same reasons the pods, lenses and streaks of ore in the endomorphosed igneous rock, as sketched in Fig. 7, could have resulted only from the passage of solutions after the consolidation of the rock.

GENESIS OF THE EMERY.

Except for the early paper by Williams, which was written at a time when contact-metamorphic processes were not well understood, the papers written on the genesis of the Peekskill emery seem to have started with a thesis based on laboratory investigation of simple systems, rather than on attempts at interpreting field observations.

Rogers based his conclusions on the results of an interesting series of experiments by Morozewicz,[18] who found that corundum, sillimanite, and cordierite will form in a melt, in which are varying proportions of alumina, silica, magnesia, and iron oxide. Rogers recognized that important differences existed between the observed mineralogical features of the Peekskill ore and the results of the laboratory experiments, but he nevertheless explained the formation of the Peekskill emery and its gangue as the result of the reaction between the basic magma and aluminous wall rock, because Morozewicz had shown that clay crucibles were

[18] Morozewicz, J., *op. cit.*

attacked at a high temperature by melts rich in MgO and low in Al_2O_3,K_2O, and Na_2O.

It is of course now common knowledge, because of the large amount of work done on ceramic products, that mullite and corundum will form in clay bricks used as furnace linings in contact with basic slags. Bowen was aware of these reactions from his study of the alumina system, and he was quick to accept Rogers' description of the Peekskill emery as an excellent field example of these laboratory experiments.

Bowen [19] compared the composite analyses of the Manhattan schist away from the igneous border, with other analyses made of it where in contact with the Cortlandt series, and he noted a striking increase in alumina and a decrease in silica and magnesia. After a technical theoretical discussion of the probable reactions, he concludes that a basic magma, such as the Peekskill norite, reacted with the schist, an old aluminous sediment, and that the emery ore is the result of the reaction of sillimanite-rich inclusions with the magma. There resulted a partial solution of such inclusions, and a recrystallization of the "surplus material" as masses rich in corundum, spinel and cordierite. The feldspathic emery and the noritic emery he regarded as inclusions approaching final disappearance in the magma. He was not thinking of an attenuated "ore magma," but of a typical basic magma of normal type.

Bowen made no attempt to reconcile field observations with his conclusions, which were based simply on the knowledge that mullite and corundum will form in an aluminous material in contact with a magnesia-rich melt.

Larsen has given attention to the interesting problem of corundum deposits in general, in observations extending over a period of years and made at a number of deposits. In a recent paper [20] he has given an exhaustive review of the literature, and advances the thesis that "plumasite bodies" (corundum-aplites) are due to

[19] Bowen, N. L., "The Behavior of Inclusions in Igneous Magmas," *Jour. Geol.*, vol. 30, pp. 551–558, 1922.

[20] Larsen, E. S., "A Hydrothermal Origin of Corundum and Albitite Bodies," Econ. Geol., vol. 23, pp. 398–433, 1928.

hydrothermal action rather than resulting from a reaction between a magma and a wall rock, with consequent de-silication of the magma. In regard to the Peekskill emery, he says:[21]

"... a study of the literature and a brief visit to the field has led me to believe that the emery deposits near Peekskill, N. Y., which are in schists near the contacts with norite and related rocks, are also hydrothermal contact-metamorphic deposits."

Since writing the above Dr. Larsen visited the region with the writers, who have thus had the benefit of his further observations, and of consultations with him, and they take this opportunity of acknowledging their appreciation thereof.

CONCLUSIONS.

The writers conclude that the Peekskill emery deposits are contact-metamorphic in origin, and were formed by gaseous or liquid emanations from the magma reservoir, which passed upward through the already solid border of the igneous mass, and into the schists, depositing the ore minerals in both the endomorphic and exomorphic zones. They base their conclusions on the following points of evidence:

1. The emery is associated with a metamorphosed schist, and a modified igneous rock, and study shows that the latter was altered by post-consolidation processes, and only at the contact, *i.e.*, it was endomorphosed. The emery occurs only with this endomorphosed igneous rock; it is not found in contact with the fresh mela-norite.

2. The associated minerals of the gangue, and the minerals of the ore itself—corundum, spinel and magnetite—are common contact-metamorphic minerals.

3. The sequence of mineralization, which began on both sides of the contact with an andesinization of the labradorite, is characteristic of contact-metamorphic processes. The sequence began with minerals known to form at high temperatures—sillimanite, cyanite, garnet, and tourmaline; and it closed with a formation of others common to lower temperature conditions—chlorite, magnetite, and sulphides.

4. The emery occurs in veins, bands, layers, lenses and pods in the schist, and in the endomorphosed igneous rock. Many of these are very thin but continuous for considerable distances, and many are found so far within the wall rock as to preclude the possibility of their formation by magmatic reaction.

[21] *Idem*, p. 429.

5. The feldspar of the emery is andesine, and not labradorite. It could not have resulted from an equilibrium between the basic norite magma and the wall rock. This observation is considered to be an especially important one, since the andesine was an early mineral in the paragenesis. As stated under 3, the andesinization of the labradorite was a post-consolidation process—yet the formation of the emery followed it. Hence the emery could not have resulted from a reaction between the basic norite magma and the wall rock. There was no basic magma within hundreds of feet at the time the emery formed, for the igneous rock had solidified at its margin.

6. Quartz and corundum occur in the same thin sections of endomorphosed igneous rock. Under any conception of the formation of the minerals by reaction and assimilation, these two minerals would not have formed separately, but would have united into an aluminum silicate.

7. Many of the minerals prove, by their very presence, an origin by emanations—the tourmaline, the rutilated quartz with cavities in which are moving bubbles, the veins of andesine and oligoclase, the chlorite, the pyrrhotite, the deuteric magnetite; and the presence of abundant apatite, zircon and some allanite in igneous rocks elsewhere without these minerals.

Bowen cited these deposits as being due to reaction because of the analogous character to the corundum-sillimanite product found by experiment. On the very basis of analogy the deposits can be called contact-metamorphic, as they have all the characteristics of such deposits, especially of other corundum-bearing deposits, as summarized by Larsen.

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THE INCIPIENT OXIDATION OF GALENA.

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Introduction.

Some experimental work[1] has been done on the oxidation of galena and much valuable information concerning its oxidation products[2] has been made available, but as yet little has been published on the initial or incipient stages of its alteration as revealed from the study of partially oxidized specimens from the outcrop. At the suggestion of Dr. F. B. Laney, of the Department of Geology at the University of Idaho, the writer began an investigation of the alteration of galena in incipient stages, using as material several hundred specimens of partially oxidized lead ore, mainly from deposits in the Coeur d'Alene district of northern Idaho. These specimens were studied in polished surfaces at the University of Chicago under the direction of Dr. E. S. Bastin, and to both men the writer is very grateful. The studies confirmed the early experimental work of Gottschalk and Buehler,[3] especially the attack of oxidizing solutions on galena in the presence of other sulphides, and in addition brought new data on the unique structures assumed by supergene covellite in replacing galena.

General Features of Alteration.

Residual grains and nodules of galena very commonly occur in the oxidized croppings of ore bodies, not because galena is resistant to the attack of oxidizing agencies for, as discussed later, it is even more susceptible to attack than admixed pyrite and chalcopyrite, but because the insoluble coating of sulphate or carbonate that is formed protects the galena and tends to keep fur-

[1] Gottschalk, V. H., and Buehler, H. A., "Oxidation of Sulphides," Econ. Geol., vol. 7, pp. 13–34, 1912.

[2] Boswell, P. F., and Blanchard, Roland, "Oxidation Products Derived from Sphalerite and Galena," Econ. Geol., vol. 22, pp. 419–453, 1927.

[3] *Op. cit.*, pp. 15–34.

ther oxygen from reaching the enclosed sulphide. The two most common and abundant secondary lead minerals are anglesite, the sulphate, and cerussite, the carbonate; both exceedingly insoluble in aqueous solutions. Of the two, anglesite is most soluble, to the extent of 42 mg. per liter of water, whereas the carbonate at the same ordinary temperatures and pressures is soluble in water to the extent of 3 mg. per liter. If the water is saturated with CO_2, the solubility of the carbonate is increased several fold, lead bicarbonate being 20.8 times as soluble as lead carbonate.[4] Nevertheless, the extreme insolubility of the two alteration products does not always prevent their removal from the outcrop that has been undergoing oxidation for a long time. Usually, the length of time required and the other conditions necessary are not ideally found in nature, and outcrops without the lead sulphate and carbonate and generally also without residual masses of galena are the exception.

Galena is oxidized in nature either as the result of air-water processes or by attack of ferric sulphate solutions. Other processes are much subordinate to these, but that of lead sulphate formation as a result of the action of copper sulphate on galena with covellite as another reaction product might be mentioned. However, this is a double decomposition reaction and not a process of oxidation. Sulphuric acid has been cited as an agent that might attack galena, but Nishihara [5] has shown that galena reacts readily with dilute sulphuric acid only because of the presence of minute quantities of included manganese sulphide. Actually, galena may be deposited in an acid environment and apparently the only effect the acid has is in preventing the hydrolysis of iron salts that might be present.

The simplest reaction and probably the dominant process in nature is the air-water process that is represented by the following equation:

$$PbS + 2O_2 \rightarrow PbSO_4$$

[4] Leitmeier, H., "Bleicarbonat, in Dollter, C.," *Handbuch der Mineral Chemie*, vol. 1, pp. 513–514, 1912.

[5] Nishihara, G. S., "The Rate of Reduction of Descending Waters by Certain Ore and Gangue Minerals," ECON. GEOL., vol. 9, p. 747, 1914.

This reaction has been demonstrated experimentally by Gottschalk and Buehler [6] and has been cited by Boswell and Blanchard [7] as checking with conditions actually found in the field, that galena may dissolve without generation of, or attack by, iron-bearing solutions, and commonly does so. In such cases, it leaves no limonite. This process receives further confirmation from the writer's studies, for in most specimens the galena has been replaced by anglesite without trace of iron salts. Limonite might be present in some, but its relations indicate that it has been borne in later and deposited on the sulphate or carbonate as a result of hydrolysis of ferric sulphate.

The oxidation of galena by ferric sulphate solutions such as may occur in the presence of admixed pyrite or chalcopyrite or from imported iron-bearing solutions unquestionably plays a rôle. This reaction has also been demonstrated experimentally by Cooke,[8] who found that a gram of galena in contact with sulphuric acid and ferric sulphate gained (by oxidation) 0.17 grams in 81 days, and without the ferric sulphate gained only 0.0714 grams.

The equation assigned to this reaction is

$$Fe_2(SO_4)_3 + PbS + 3O + H_2O \rightarrow PbSO_4 + 2FeSO_4 + H_2SO_4.$$

Boswell and Blanchard [9] also discuss the importance of this reaction in nature and indicate that, if limonite is left with the products, it signifies that the sulphides carried admixed pyrite, or other iron-yielding minerals, or that the iron-bearing solutions were imported during oxidation of the lead mineral. They explain further that excess ferric sulphate left over from the oxidation of the pyrite could react with lead carbonate, forming lead sulphate and ferric hydroxide (from which limonite would result), and they consider this the most probable and common source for limonites derived from lead minerals. Such relations

[6] *Op. cit.* pp. 13–31.

[7] *Op. cit.* pp. 445, 451.

[8] Cooke, H. C., "Secondary Enrichment of Silver Ores," *Jour. Geol.*, vol. 21, p. 11, 1913.

[9] *Op. cit.*, pp. 447–450, 451–452.

were also attained in the ores studied by the writer. In most cases the limonite associated with anglesite or cerussite was deposited from later solutions that reacted with the carbonate, although in some it is clearly the reaction product of ferric sulphate with galena and the result of hydrolysis of ferric sulphate from the reoxidized ferrous sulphate. The reaction showing the attack on lead carbonate by ferric sulphate solutions may be expressed as follows:

$$3PbCO_3 + Fe_2(SO_4)_3 + 6H_2O \rightarrow 3PbSO_4 + 2Fe(OH)_3 + 3H_2O + 3CO_2.$$

Another feature of note is that the galena first oxidizes to sulphate and apparently any carbonate that might form does so by replacement of anglesite and never of galena directly. Such conclusions were obtained in all the specimens studied and are in line with similar conclusions made by Wang,[10] who found in regard to the alteration of sphalerite, that carbonate never replaces sphalerite directly.

Additional features of general interest in the oxidation of galena beyond the incipient stage are ably presented by Boswell and Blanchard,[11] who, from the oxidation products remaining in place after the cropping has been leached and no longer carries essential sulphates, carbonates, or other oxidized minerals of the original sulphides, use the physical characteristics of the limonites themselves for interpretation of the former presence of galena.

Incipient Stages of Alteration.

Replacement of Galena by Anglesite.—Replacement of galena by anglesite may take place through air-water processes, by reaction with ferric sulphate, or by reaction with copper sulphate, as discussed in the preceding section, but only results of the first two processes will be considered here. Whichever process was active, the result in the incipient stage is much the same. The early alteration of galena or its replacement by anglesite is mainly

[10] Wang, Y. T., "The Formation of the Oxidized Ores of Zinc from the Sulphide," *Trans. Amer. Inst. Min. Eng.*, 1915, p. 1001.

[11] *Op. cit.*, pp. 419–453.

directed or controlled by the cleavage of the galena, the usual relations being illustrated in Fig. 1. In general, certain cleavage directions seem more susceptible to diffusion of oxidizing solutions than do others and these cleavages do not always appear the most pronounced in polished surfaces. Usually, these minute veinlets ramify in two directions. The replacement border is extremely irregular, with saw-like edges. As the veinlets continue to enlarge, the borders become more and more irregular (on a larger scale) and as replacement advances more rapidly in some directions than in others, masses of the galena eventually are isolated in the anglesite. These isolated masses then tend to assume more or less nodular forms as the corners are removed by the attacking solutions. Usually, even in the smallest veinlets, the anglesite abounds with minute inclusions of the galena. The structure of the replacing anglesite veinlets illustrated in Fig 1 is especially typical of the air-water process, but similar tendencies may be noted when alteration has been at least in part by ferric sulphate solutions.

Of more than usual interest in this replacement is the apparent absence of volume change which the reaction demands. The anglesite is usually dense, especially when limonite is lacking, homogeneous, and forms a knife-like contact with the galena. No narrow spongy zone is visable except where the sulphate has been later attacked by iron or carbonate solutions. It is probable that the excess sulphate demanded by the chemical equation has been deposited elsewhere as a result of electrolysis, which might explain the presence of fine prismatic crystals of anglesite in pores of many partially oxidized specimens.

Although air-water processes play the major rôle during incipient stages of oxidation, nevertheless in some cases ferric sulphate solutions also have been active. Some veinlets have central zones or cores of limonitic anglesite as illustrated in Fig. 2. It is always difficult to determine whether the limonite in such cases has been formed as a result of hydrolysis of the attacking ferric sulphate solutions or brought in later. Where the former reaction can be determined, a dense white zone of anglesite between

the limonitic material and the galena is always shown, suggesting that the limonite could not be deposited near the galena because of the ferrous sulphate produced during the reaction. Ferrous sulphate does not hydrolyze, but in diffusing outward from the galena would encounter oxidizing conditions and be reoxidized to the ferric state. Then, probably because of decrease in acidity of the solution, due to neutralization or dilution, it would hydrolize, yielding limonite. Generally the limonitic zone becomes increasingly porous outward, especially when the incipient stage of alteration no longer exists, but the increase in porosity usually implies the attack of later agencies and the solution of the anglesite. As the ferric sulphate solutions are probably always accompanied by dissolved oxygen, it would be difficult in any event to determine how much of the incipient alteration is due to dissolved oxygen and how much to ferric sulphate. The specimens examined suggest that the oxygen invariably takes the lead or causes most of the alteration. As a result of the mutual attack of both kinds of oxidizing agents, the limonitic zones often assume peculiar string-like or narrow ribbon-like bands, producing a more or less cell-like structure in the oxidized products.

Oxidation in the Presence of Admixed Pyrite and Chalcopyrite.—When either pyrite or chalcopyrite are admixed with the galena, oxidation of the latter begins before the others and is apparently hastened by their presence. The effect of these minerals and other sulphides on the rate of oxidation was noted in laboratory experiments by Gottschalk and Buehler,[12] who found that pyrite or marcasite when mixed with other sulphides caused a more rapid oxidation of the second sulphide. In the presence of equal amounts of pyrite, the galena, sphalerite, covellite, and enargite were found to oxidize from 8 to 20 times faster than when treated alone, and in each case the oxidation of the pyrite was greatly retarded. They explained that the acceleration of the reaction was due to electric currents generated by contact of minerals of different potentials, the electrical current flowing from the mineral having the higher potential, as a result of which

[12] *Op. cit.*, pp. 15–34.

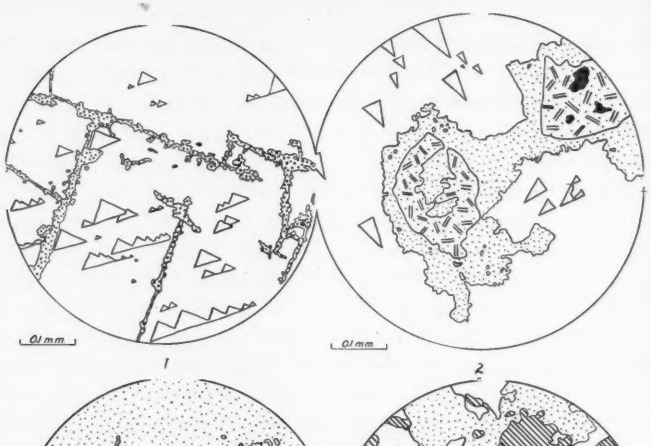

FIG. 1. A characteristic structure assumed by anglesite (stippled) in replacing galena during incipient stages of oxidation. Note how the veinlet pattern is controlled or directed by the cleavage of the galena (shown by the triangular pits). Such veinlets give no indication of volume change related to the oxidation.

FIG. 2. A structure developed when part of galena replacement by anglesite has been by reaction with ferric sulphate solutions. The limonite, produced by hydrolysis of the ferric sulphate, forms the inner core of the anglesite seam (stippled). Much anglesite also occurs in the limonitic zones (stippled and pattern of short parallel lines).

FIG. 3. Crystals of pyrite (ruled) entirely enclosed in the anglesite zone (stippled) and showing not the least sign of attack by oxidizing solutions, except for one crystal in the lower right corner. Only minor areas of unoxidized galena (white) remain in the section.

FIG. 4. Grains of chalcopyrite (ruled) surrounded by anglesite (stippled) or in contact with both anglesite and galena (white) showing no sign of attack by the solutions that altered the galena.

the mineral with the lower potential was dissolved or attacked more rapidly, and the one of higher potential was protected from oxidation and solution. That this behavior was actually due to potential differences and electrical currents was ably demonstrated by them by taking a piece of galena and one of pyrite, attaching a copper wire to each and forming a circuit by placing a galvonometer between the free ends of the wires, then placing the minerals in distilled water but without permitting the copper wires to touch the water. A current of five microamperes was found to flow through the circuit and in the course of a few days, the circuit being maintained, the galena became coated with lead sulphate, while a similar piece of galena placed in the same solution, but not in the same circuit, remained perfectly bright. The pyrite used in the cell showed no evidence of oxidation. They thus obtained the potential of a number of sulphides by means of the Ostwald potentiometer while the minerals were immersed in distilled water, each measured against copper wire. Subsequently Wells [13] studied the electrical activity in ore deposits and found that nearly all metallic sulphides except sphalerite are good conductors of electricity, while nearly all sulphates, carbonates, and silicates are poor conductors. Their potentials vary greatly with variations in the solutions employed, and as chemical activity continues the salts that are formed by decomposition of the minerals will themselves cause electrical action to change. The acid and oxidizing solutions in general gave the highest potentials and the alkaline and reducing solutions the lowest.

That such conditions and relations actually obtain in nature have every confirmation in the specimens studied by the writer. In most sections, either pyrite or chalcopyrite, and usually both, occur in minor amounts in the galena and in the anglesite, and curiously those in the bands of anglesite show not the least evidence of alteration unless the band of sulphate is wide and the pyrite or chalcopyrite some distance removed from the galena. These relations are illustrated in Fig. 3 for pyrite and in Fig. 4 for chalcopyrite. In the former the crystals are entirely sur-

[13] Wells, R. C., "Electrical Activity in Ore Deposits," U. S. Geol. Survey Bull. 584, p. 79, 1914.

rounded by anglesite, yet they maintain sharp crystal borders and the anglesite shows no sign of iron staining. In Fig. 4 the chalcopyrite maintains the same outlines against the anglesite as against the galena and shows no evidence of chemical attack, which would be readily indicated by patches of covellite had solution of the chalcopyrite occurred. These relations persist in all specimens, except, as mentioned before, where the pyrite or chalcopyrite is some distance from the galena, in which case oxidation goes on independently.

In general, the galena is most extensively altered in zones richest in pyrite or chalcopyrite. The anglesite zone in most sections tends to swell or enlarge about the admixed sulphides, in some places very widely. These relations infer that the presence of pyrite and chalcopyrite actually increases the rate of oxidation of galena while the other sulphides at the same time are actually retarded in their oxidation. This is a corroborative check of the experimental work of Gottschalk and Buehler. They also determined the potential of pyrite to be + 0.18 volts, of chalcopyrite + 0.18 to + 0.30 volts, and of galena + 0.15 volts and, as mentioned earlier, the mineral with the higher potential is protected from oxidation and solution and the one with lower is attacked more rapidly. Obviously, as the zone of anglesite intervening between the galena and pyrite or chalcopyrite becomes increasingly large the electrical forces between the galena and either sulphide become decreasingly weaker. Finally the cell is destroyed and the pyrite and chalcopyrite are oxidized independently as evidenced by empty or limonite-filled cavities and in the case of chalcopyrite by covellite patches in the anglesite or on the galena border.

Attempt was also made to study the effect of admixture of tetrahedrite, sphalerite, and other minerals on the oxidation of galena, but the proper specimens were not available. Sphalerite was present in many specimens, always in contact with galena, but never with anglesite. Apparently the sphalerite is much more susceptible to attack than galena; in fact, so highly susceptible that if galena shows any evidence of oxidation, the sphalerite has already been oxidized and because of the solubility of its sulphate

transported elsewhere. A recourse to oxidation potentials shows that sphalerite has a very negative potential of —0.2 to —0.4 volts. Thus galena with its higher positive potential would be the mineral protected from oxidation and the sphalerite the one more readily attacked in an admixture of these two minerals.

Replacement of Galena by Covellite.—In many of the specimens examined, secondary covellite after galena is a common though by no means an abundant mineral. The equation which expresses the reaction between galena and copper sulphate solutions may be written as follows:

$$PbS + CuSO_4 \rightarrow PbSO_4 + CuS.$$

The covellite produced either forms a border on the galena or occurs in patches in the anglesite. Sources of the copper solutions were determined in some sections as from scattered grains of chalcopyrite that finally had been attacked by oxidation, but in others the copper sulphate was brought in from the outside. Apparently, in all cases the galena acts as a ready precipitant of copper sulphate, but covellite generally is not found in large amounts with partially oxidized galena because of the general lack of association of copper and lead minerals. Further, the insolubility of the anglesite tends to prevent access of fresh galena to attacks by copper sulphate solutions, thereby also limiting the covellite formation.

Curiously, the covellite assumes some very remarkable structures in replacing galena, taking many shapes and forms, but resolving itself into four main characteristic groups. For convenience, these may be listed as (1) "regular" borders, (2) "botryoidal" structures, (3) "stranded" borders, and (4) "platy" structures. Several of these structures may occur in the same specimen and usually do, especially the "regular" and "stranded." These covellite structures rarely follow cleavage lines.

The "regular" structure is that illustrated in Fig. 5, wherein the covellite forms a narrow band on the galena, maintaining a more or less constant width. It might perhaps as well be called

a ribbon structure. The band usually develops parallel to cleavage directions in the galena and is usually accompanied by an inner casing of anglesite of about equal width. The structure is best displayed when the altering solution has been primarily copper sulphate. Strangely, this kind of structure is the least common. More generally the covellite forms irregular and disconnected patches or borders on the galena as illustrated in Fig. 6. Some patches enlarge greatly within the galena, their enlargement being irrespective of galena cleavages. Apparently this structure is obtained where copper sulphate is not uniformly concentrated throughout the solutions, typical, perhaps, of cases in which disseminated chalcopyrite or tetrahedrite grains in the galena are the source of copper.

The second and generally the most common in the specimens examined is the "botryoidal" structure illustrated in Fig. 7. In this the covellite and anglesite are more or less zonally arranged within elliptical to spherical bodies at the margin of or within the galena. The globular or botryoidal outline of the replacing mineral is especially striking, but difficult to portray by camera lucida drawing. The margin of these bodies is very finely irregular, composed of minute saw-like projections, probably minute covellite laths. Within are usually alternating concentric rings or bands of covellite and anglesite, the covellite predominating, as it forms much wider bands. The inner core is generally anglesite with minor seams or partial rings of covellite arranged concentrically. Many bodies, however, are composed wholly of covellite. The form of these masses and their internal arrangement suggest a gel or colloidal replacement.

The third structure described as "stranded" borders is very common in most sections. In this, covellite forms bands or ribbons or more usually patches within the anglesite, always separated from the galena. This characteristic structure is illustrated in Fig. 8 and results from any of the other structural types when they become separated from the galena or stranded in the anglesite. In Fig. 8, the covellite bands apparently were directed by the original galena cleavage and subsequently have become detached from the galena. This relationship suggests that, fol-

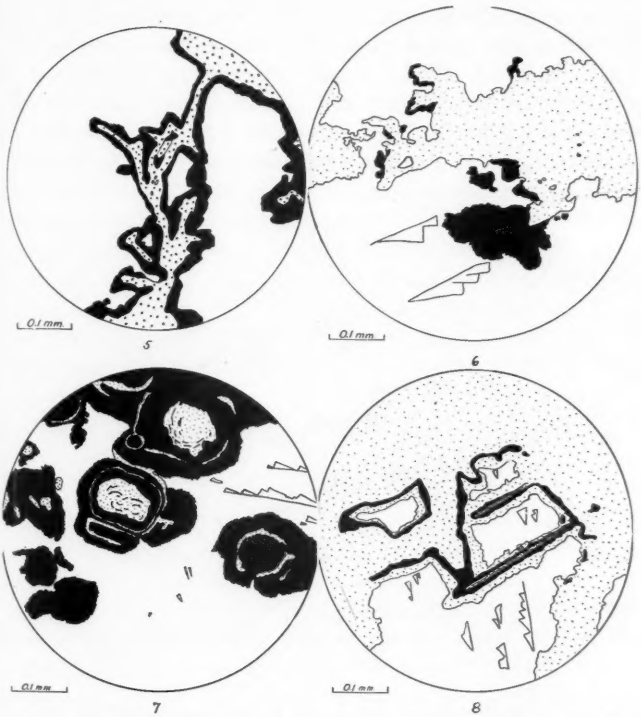

FIG. 5. "Regular" border occasionally formed when galena is replaced by covellite. The covellite is indicated by the black bands, the anglesite by stippling, and the galena by its lack of markings.

FIG. 6. A more common occurrence of the "regular" border where the covellite tends to form as irregular and disconnected patches or partial borders instead of as continuous bands. Markings the same as in Fig. 5.

FIG. 7. "Botryoidal" structure assumed by covellite in replacing galena. Such structures are common in many specimens and in their form and internal arrangement suggest gel or colloidal replacements. Markings the same as in Fig. 5.

FIG. 8. "Stranded" covellite borders, a common structure in most specimens, suggestive of stranding of the former border by further oxidation of the galena. Markings the same as in Fig. 5.

lowing covellite deposition, the oxidation of the galena continued after all copper had been precipitated or that solutions with oxygen were introduced later and continued the oxidation of the galena without affecting the covellite. In either case, oxidation of the galena continued along the same fronts. To account for the stranding of the covellite and its apparent resistance to oxidation, it is only necessary to call in the electric activity principle. Gottschalk and Buehler [14] found that covellite has a potential of

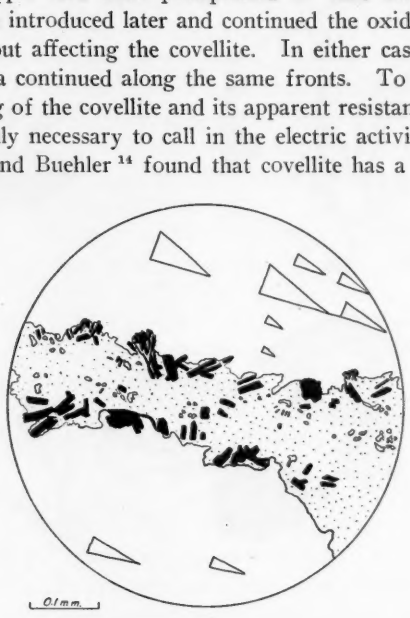

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FIG. 9. "Platy" covellite structure. Note how the covellite assumes its own crystal form in replacing the galena and how many of these crystals have been stranded in the anglesite. Markings same as in Fig. 5.

+ 0.20 volts, a potential higher than pyrite, and applying this principle, the covellite should be protected from attack while the galena with its lower potential should be more rapidly oxidized. Such an explanation would account for the patches of covellite where found within the anglesite. Obviously, as the intervening band of anglesite increases, the cell eventually is destroyed and the covellite attacked independently of the galena, the copper again taken into solution, and again deposited on the border of the galena. This process might be repeated indefinitely.

[14] *Op. cit.*,

The final structure is the "platy" structure illustrated in Fig. 9. This one is unusual and was found in a specimen from the Mowry Mine, Patagonia district, Arizona.[15] The specimen is from the lower oxidized zone of a galena-rich ore formed by replacement of Pennsylvanian limestone. The primary minerals are galena, pyrite, arsenopyrite, tennantite (?), quartz, and calcite, and the secondary covellite was derived from solutions descending from the upper oxidized zone. In replacing the galena, the covellite assumes its own characteristic tabular or platy form. Some plates penetrate the galena whereas others are stranded in the anglesite. This type of replacement seems rare, although there were suggestions of it in some of the other sections, but in crystals so minute as not to be entirely distinguished. Thus the tendency of covellite to assume its own crystal form in replacing galena is another of its peculiarities.

Summary.

Study of incipient stages of alteration of galena indicates that the galena is more generally oxidized by air-water processes than by solutions of ferric sulphate. Much of the limonite, commonly associated with the oxidation products of galena, represents a later addition from external sources. The first product obtained by oxidation of galena is always anglesite. In the earliest stages, the replacement of galena by anglesite is mainly directed and controlled by cleavage directions in the galena. Of more than usual interest is the apparent absence of volume change from galena to anglesite.

With an admixture of pyrite and chalcopyrite, the oxidation of galena becomes more complex. Due to the formation of electrolytic cells between the various sulphides in contact with the oxidizing solutions, the attack on galena is greatly aided while the oxidation of pyrite or chalcopyrite is very much retarded. On this basis the galena oxidizes earlier and with greater ease than most other sulphides, except sphalerite. Galena remains in the

[15] The specimen and details of its occurrence were kindly furnished by Dr. E. S. Bastin of the University of Chicago.

oxidized croppings only because the coating of insoluble alteration products prevents ready access by fresh oxidizing solutions.

Covellite is a common though not abundant mineral formed by replacement of galena. It, curiously, assumes some very interesting and diverse structures in its replacement, but four main types of structures may be recognized. For convenience, these may be listed as (1) "regular" borders, (2) "botryoidal" structures, (3) "stranded" borders, and (4) "platy" structures, each referring to a relation the covellite bears to the galena.

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EDITORIAL

OFFICIAL RECORDING OF MINING GEOLOGY

THE mining laws of Sweden impose upon the operator of each mine the duty to have the mine surveyed not only geometrically but also geologically, and to deposit a copy of the map in a certain government bureau. Only open cuts that can be entirely inspected from the surface are exempted. These regulations are probably unique, and it may perhaps interest the readers of this journal to hear some details about them, and how they work out in practice. A good and rather detailed account was given in English by Professor W. Petersson twenty years ago, in the Proceedings of the Eleventh International Geological Congress, meeting in Stockholm in 1910. These lines will serve to call attention to Petersson's paper, and give those who do not have access to it some idea of the subject.[1]

Mine mapping in Sweden was started in consequence of an edict by King Gustavus Adolphus in 1628, but it was not until the following century that geological conditions also began to be indicated on the maps. The present regulations date mainly from 1884 and 1885, supplemented by some later modifications. A mine surveyor's license is given only to persons sufficiently experienced in geology as well as in ordinary surveying. The standard scale of the maps is 1 : 800 for metalliferous mines, and 1 : 1500 for coal mines. The following details apply only to the metalliferous mines.

A mine map consists of a series of horizontal sections of the mine, spaced so closely as to show each level of any importance (not merely haulage levels, etc.), with a vertical interval as a rule of about 10 meters, somewhat varying according to the geological regularity of the deposit, the mining method employed, and other

[1] Some modifications have recently been introduced, but they do not affect the basic principles, therefore the paper in question is still useful.

local conditions. There is also a series of vertical cross-sections, and a projection of the whole mine on a vertical plane along its longer axis, all on the same scale. Outlines of workings are shown in black, openings continuing down to the next sheet are indicated by hachures, and the depth below a certain datum is marked at numerous points. The geological conditions are shown in colors, all openings and their walls being thus mapped, but no constructions or combinations outside these known areas are permitted. A standard color scheme is followed, to secure as much uniformity as possible and to facilitate comparisons, but certain possibilities of modifying and supplementing are left open. Observed faults are, of course, duly indicated, and structural features in general are shown by the usual symbols. A map of an imaginary mine has been published to serve as a guide for the practical details, choice of colors, etc. A complete copy of each map is deposited in the office of the government mining bureau, and all new developments are to be reported within a certain time margin, to be introduced on this copy.

Obviously, not every map can be of the same quality, for the geological ability of the surveyors is not always the same, and the natural phenomena they are to record may present greater difficulties in one mine than in another. On the whole, however, the system may be said to have worked out very well indeed. Each mine owner has been forced, for his own good, to keep a map record not only of the progress of the underground work, but also of the geological conditions encountered, and with comparatively little additional trouble for the mining administrations the community is supplied with a map collection of great practical value. It is thus made possible for officers of the government institutions to get information about the geology in now inaccessible parts of a mine. For the work of the Geological Survey, these map files are particularly useful. The same opportunities for study are also open to others, and this fact is not the least important feature of this arrangement. For the question may arise of re-opening an abandoned mine at a time when, perhaps, the old administration is dispersed and no private records can be ob-

tained to give information about the extension of the old workings and the geological structure of the deposit. Also a mine is not, according to the mining law of the country, a private property in an ordinary sense. The discoverer of a metalliferous deposit, and the land owner, are each entitled to half ownership, but they have this right only so long as they fulfil certain conditions. If the owners forfeit their rights or otherwise give up the mine, it is imperative that those who may be willing to make a new attempt shall have access to as much as possible of the experience already obtained. A mine, although a private enterprise, is of great importance to the community as a producer of opportunities of employment and as a taxpayer, and the community is therefore entitled to a certain control over it.

The practicability of the Swedish mine-mapping law rests upon certain favorable conditions, such as the comparatively small variation in the geological character of the deposits, and it can hardly be applied in exactly the same form in more than a few other countries. The general principles underlying it, however, are capable of a much wider application. It would be of interest to hear whether they have been put in practice elsewhere.

PER GEIJER.

DISCUSSION AND COMMUNICATIONS

SUPERGENE CASSITERITE IN TIN VEINS.[1]

Sir: I read with much interest the paper by Singewald on the possibilities of the formation of supergene cassiterite in tin deposits and the ensuing discussion between Koeberlin and Singewald. Inasmuch as during recent years (1924–1928) I have had opportunity to examine most of the Bolivian tin deposits, to collect extensive material from them, and to study that material microscopically, I venture a contribution to the above subject in the form of a brief resumé of the results of my investigations.

The crystal form of cassiterite varies with the conditions under which it is deposited. I can distinguish the following types in Bolivia:

Pegmatitic Form: Mostly simple ditetragonal pyramids, with pronounced zonal growths; but also twins after the Zinnwald law (Fabulosa Mine).

Pneumatolytic Form: Zinnwald twins predominant, occasionally clear and colorless, but frequently dark through presence of iron (Uncia). Here belong the occurrences of Araca, Llallagua, etc. Rarer are simple, thick, tetragonal prisms with pyramidal faces (Cornwall type from the San Luis vein at Monte Blanco and from Chualla). With decreasing temperature the crystals are smaller.

Hydrothermal Form: Small individuals, mainly the Cornwall type (Jatuncaca vein at Colcha, Colquiri). With decreasing temperature the crystals approach the needle tin type. Most of the cassiterite has been deposited in colloidal state and frequently in radial, fibrous to finely crystalline masses, with simple needle crystals in druses (Oruro, Potosí, Chocaya).

To obtain criteria to distinguish between hypogene and super-

[1] Translated from the German manuscript by Joseph T. Singewald, Jr.

gene cassiterite, specimens of undoubted supergene origin were examined microscopically. They show generally straw-yellow color, fine porous texture, finely crystalline structure, and marked contamination by anglesite, smithsonite, and limonite. The differentiation of hypogene and supergene cassiterite under the microscope is not easy and requires much practice.

According to my experience, meteoric waters are not capable of dissolving cassiterite since they (especially in Bolivia) are always acid. Alkaline surface waters are theoretically capable of dissolving cassiterite. But such waters are not present in Bolivia. I could observe frequently, however, that cassiterite had been dissolved by ascending thermal waters and presumably redeposited nearer the surface as cassiterite in the form of wood or kidney tin or as stannite. These processes appear to play an important rôle in Bolivia. Such phenomena I observed at Uncia-Llallagua, Chualla, and in other places. Perhaps the young, near-surface wood tin deposits, as for example in the andesite flows south of Colquechaca, may be explained in this way. In order to dissolve cassiterite, the thermal waters must have been alkaline.

All the supergene cassiterite observed by me was derived from the weathering of sulphostannates. It should be pointed out that they, especially stannite, play a much greater rôle in the Bolivian deposits than has been recognized. Stannite occurs microscopically in practically all the hydrothermal tin deposits of central and southern Bolivia, in some as the principal ore (Pomabamba vein of the El Salvador mine, at Pasña). It is lacking only in the pneumatolytic deposits (as, for example, Caracoles). Stannite is very common in the Uncia-Llallagua veins, generally microscopic, especially in the upper levels. Franckeite I found in 14 deposits of central and southern Bolivia, in two of them as an important ore (Porvenir and Maria Francisca, near Huanuni); cylindrite in 3, and in one of them (Poopo) as an important ore; zincteallite as an important ore in two mines (Monserrat at Poopo and Carguaicollo). All these sulphostannates yield to weathering near the surface and give rise to the formation of supergene cassiterite.

Stannite weathers less readily than pyrrhotite but more readily

than pyrite. Its decomposition results in earthy black covellite and cassiterite. Further weathering leaches out the covellite which goes into solution as the sulphate. There remains a porous, pulverulent cassiterite (not wood tin). Descending stanniferous solutions do not result. The cassiterite remains where it formed. Therefore, secondary enrichment can not take place in this process. The phenomena accompanying the weathering of stannite had not previously been studied because the oxidation zone of the stannite-rich deposits (Oruro, Llallagua, Colquechaca, Chocaya) has long since been worked out and is no longer accessible. I had an especially favorable opportunity to study them in the newly discovered Vila Apacheta mine at Chayanta.

The deposits with franckeite and cylindrite show no marked oxidation phenomena at the outcrop. The Porvenir vein at the tunnel level, 40 m. below the outcrop, is quite unoxidized. Zinc-teallite, on the other hand, because of its marked basal cleavage, breaks down more readily. Therefore, we find in the outcrop of the Monserrat and Carguaicolla deposits larger quantities of supergene cassiterite. I recently described the latter deposit.[2] Here also there has been no downward migration of the cassiterite. The tin content of the vein near the outcrop where the teallite has been completely changed to cassiterite is the same as at greater depths.

The occurrence of supergene cassiterite plays a very minor rôle economically; it has not brought about secondary enrichment of the tin content.

In conclusion the danger should be pointed out that results from a one-sided consideration of these processes on the part of the field geologist. A clear understanding of them can be attained only after a deposit has been thoroughly studied and specimens collected from various depths and microscopically investigated. My own ideas arrived at as a field geologist in Bolivia had to be revised in the light of the results of my subsequent microscopic investigations.

FRIEDRICH AHLFELD.

MARBURG, GERMANY.

[2] Die Zinnerzlagerstätte von Carguaicolla (Bolivien). *Zeit. f. prakt. Geol.*, vol. 37, pp. 216–219, 1929.

THE IRON DEPOSITS OF THE SIERRA DE IMATACA, VENEZUELA.

Sir: In the January-February, 1930, number of ECONOMIC GEOLOGY, Guillermo Zuloaga presents a communication concerning this subject which is of interest to me in view of the recently published paper on the Pao iron ore deposits.[1]

Mr. Zuloaga refers to the Pao deposits, which are the westernmost of the iron ore deposits thus far discovered in the Sierra de Imataca, and indicates that he has made studies of a broader range of deposits in the region and more thorough laboratory studies than I was able to make. It is therefore of interest to find that certain conclusions in the two papers are very similar, for instance, that the richer iron ore deposits are closely related to igneous intrusives, and that the granite appears to be intrusive into the banded iron-bearing quartzites of the region rather than to belong to the older basement complex of the Guayana Highlands.

Mr. Zuloaga states that the geology as well as the character of the Sierra de Imataca ores seem to be remarkably similar to those features of the iron ore deposits of the Province of Minas Geraes, Brazil, as described by Harder and Chamberlin, and expresses the opinion that one theory of origin may apply to the ores of the two regions. My paper points out certain similarities in the Pao and Minas Geraes ores and also several dissimilarities. The ores of Minas Geraes are believed by Harder and his associates, who have studied them thoroughly, to have originated as ferruginous sedimentary beds, and there is a preponderance of evidence to support their conclusions. From what I have seen of the ores in the two fields, I am rather dubious about ascribing an identical history to their formation because the dissimilarities as detailed in the paper cited above are sufficient to indicate a more complex history for the concentrations of practically pure hematite and magnetite which constitute the striking features of the Pao deposits.

Further laboratory studies of the ores from both fields would

[1] Burchard, E. F., "The Pao Deposits of Iron Ore in the State of Bolivar, Venezuela," *A. I. M. M. E.*, Tech. Pub. 295 (issued January, 1930).

be useful, particularly as to the relations of hematite and magnetite. Gruner [2] has found through experimental work with hot water at temperatures between 150° and 300° C. that ferrous iron minerals are oxidized in the absence of air if the hydrogen liberated in the reaction can escape readily and that practically all iron is precipitated in this reaction, while very large quantities of silica, including quartzites, are taken into solution. He believes that hot waters rich in sodium, magnesium, calcium, and iron from intermediate or basic magmas could therefore produce replacement deposits of the type of the Pao, Venezuela, and Vermilion, Minnesota, ore bodies.

There are evidences of replacement in the Pao deposits, for, although the transition from banded siliceous ferruginous rock to practically pure iron oxide is usually rather abrupt, it is generally possible to detect a faint banding in the rich ore which may have been inherited from the earlier rock. Doctor Gruner's work seems to confirm the general conclusions reached by Mr. Zuloaga and myself as to the influence of magmatic solutions and to give a definite idea as to the nature of certain reactions.

ERNEST F. BURCHARD.

U. S. GEOLOGICAL SURVEY,
WASHINGTON, D. C.

[2] Gruner, J. W., Department of Geology, University of Minnesota, Minneapolis: Personal communication to the writer, March 8, 1930.

REVIEWS

Mining of Alluvial Deposits by Dredging and Hydraulicking. By W. E. THORNE, and A. W. HOOKE. Pp. xiv + 171. Illustrated. Mining Publications, Ltd., London, 1929. Price, 20 s.

A few years ago there was noticed in these pages[1] a little book by Thorne on the testing of alluvials for gold, diamond or tin. The author of that book has now offered another treatise on the practical methods of extracting the values from 'alluvial propositions by dredging and hydraulicking.' The book is devoted to the practice in drilling and working placer and alluvial deposits and other deposits of a similar character. It describes the tools used and the methods employed in dredging and hydraulicking, and discusses the processes upon which these methods depend for their success. A large portion of the book deals with the movement of water in flumes, ditches and pipes and its capacity for moving debris. This constitutes the first part of the volume. The second part deals with dredging by pumps, shovels, buckets and drag lines. Specifications for the parts of an efficient dredge are given and the factors that make for success under different conditions are briefly mentioned. The book is illustrated by numerous figures and scale drawings, and in an appendix are some convenient tables that should be of value to the engineer in charge of operations.

W. S. BAYLEY.

College Text Book of Geology—Part II, Historical Geology. By T. C. CHAMBERLIN AND R. D. SALISBURY. Rewritten and revised by ROLLIN T. CHAMBERLIN AND PAUL MACCLINTOCK. Pp. 878 + 28 of index; figs. 252; colored plates. Henry Holt & Co., New York, 1930. Price, $3.75.

This well-known text has been thoroughly revised and largely rewritten. The new volume is simplified and shortened. The general treatment is the same as previously but newer material has been incorporated. Many new illustrations are noted, particularly the use of paleogeographic maps which indicate the known outcrops. A striking innovation is the use of a few autochrome plates but the results are not happy; detail has been sacrificed for a few colors on a fuzzy background.

[1] ECON. GEOL., vol. 21, p. 515, 1926.

Clear pictures in black and white would illustrate the geological features better, and that is what is desired in an elementary text.

The book is highly readable, clear, well illustrated and printed. It should keep up the traditions of the Chamberlin-Salisbury textbooks and find a well-deserved place in the class room.

ALAN BATEMAN.

Oil Field Maps: Oil Fields and Pipe Lines of the United States, scale about 46 miles to the inch; **Principal Oil Fields of the World,** scale 1:22,000,000. Compiled by O. W. WELLS. Internl. Map Co. Inc., 90 West St., New York, 1930.

The first of these maps shows, in two sheets, the oil fields and pipe lines of the United States in colors. The pool outlines and names are given. There are 15 insets around the margins to show important areas in greater detail.

Similarly, the map of the world, in two sheets, gives the outlines of the important pools and their names. There are 7 marginal insets of important sections.

These maps represent an extremely careful compilation of widely scattered data and are invaluable to all interested in the petroleum industry.

Mining Methods. By CHARLES A. MITKE. Pp. 195, figs. 63. McGraw-Hill Book Co., New York, 1930. Price, $3.00.

This much needed book, printed in the excellent McGraw-Hill manner, brings together descriptions of modern metal mining methods. More than one hundred and fifty different so-called stoping methods have been reduced to a simple classification according to fundamental principles. It is obvious throughout that the author presents firsthand knowledge of most of the methods. It is a necessary book for every mining man's shelf.

Elements of Optical Mineralogy: an Introduction to Microscopic Petrography. By N. H. WINCHELL and A. N. WINCHELL. 2d edition. Part III, Determinative Tables. Pp. 204, pl. 3. John Wiley & Sons, New York, 1929 (Dec.). Price, $4.50.

The determinative tables occupy most of the book. They are: I, Opaque Minerals; II, Birefringence of Minerals; III, Color of Minerals; IV–*A*, Refringence of Isotropic Minerals; IV–*B*, Refringence of Anisotropic Minerals; V, Dispersion of Minerals. By means of these tables individual minerals may be determined. They are invaluable to every one who uses the petrographic microscope.

BOOKS RECEIVED

By

C. B. Hunt.

Methodes geophysicos applicados ás fundacoes de barragens. By E. P. de Oliviera. Pp. 9, illus. 5. Serv. Geol. e Min. do Brasil, 1929.

Aspectos econominos da Mica. By D. Guimaraes. Pp. 34, pl. 1. Serv. Geol. e Min. do Brasil, Bol. 43, 1929.

Jazidas de Minerios de Chumbo (lead deposits). By G. de Paiva. Pp. 25, pl. 10. Serv. Geol. e Min. do Brasil, Bol. 42, 1929.

A Industria do petroleo; Relatorio da viagem de estudos aos Estados Unidos Mexicanos. By G. de Faria Alvim. Pp. 66, pl. 73. Serv. Geol. e Min. do Brasil, Bol. 41, 1929.

Valle do Rio Negro: Physiographia e geologia. By G. de Paiva. Pp. 62, pl. 30, maps 4. Serv. Geol. e Min. do Brasil, Bol. 40, 1929.

La Mariupolite et ses parents. By J. Morozewicz. Pp. 130, pl. 11. Trav. du Serv. Geol. de Pologne, vol. II, liv. 3, 1929. (In Polish and French).

Oil and Gas in Oklahoma, Love and Marshall Counties. By F. M. Bullard and J. S. Redfield. Pp. 30, figs. 5, pl. 1. Okla. Geol. Serv. Bull. 40–OO, March, 1930. Structure, stratigraphy, oil and gas development.

The Geology of Ceylon. By Frank Dawson Adams. Pp. 86, figs. 2, pl. 9, geologic map of Ceylon. Can. Jour. Research, 1929. Ceylon is composed mostly of crystalline rocks with just a few Tertiary sediments locally developed. The petrography and structure of these rocks is fully discussed; 30 analyses of igneous and sedimentary rocks are included, with norms and modes. Three distinct physiographic planes of denudation are recognized. Bibliography of 70 references.

Surface Water Supply of the United States, 1925; Part VI, Missouri River Basin. Pp. 252, fig. 1. U. S. Geol. Surv. Water Supply Paper 606, 1930. Price, 30 cts.

Geology and Water Resources of the Mokelumne Area, California. By H. T. Stearns, T. W. Robinson, and G. H. Taylor. Pp. 402, figs. 33, pl. 21, maps 6. U. S. Geol. Surv. Water Supply Paper 619, 1930. Price, $1.25.

Surface Water Supply of the United States, 1926; Part IV, St. Lawrence River Basin. Pp. 163, fig. 1. U. S. Geol. Surv. Water Supply Paper 624. Price, 20 cts. **Part V,** Hudson Bay and Upper Mississippi River Basins. Pp. 170, fig. 1. U. S. Geol. Surv. Water Supply Paper 625. Price, 20 cts. **Part VI,** Missouri River Basin. Pp. 228, fig. 1. U. S. Geol. Surv. Water Supply Paper 626, 1930. Price, 25 cts. **Part VII,** Lower Mississippi River Basin. Pp. 98, fig. 1. U. S. Geol. Surv. Water Supply Paper 627, 1930. Price, 15 cts.

Water Resources of the Umpqua River and its Tributaries, Oregon. By B. E. JONES AND H. T. STEARNS. Pp. 110, pl. 10, figs. 22. U. S. Geol. Surv. Water Supply Paper 636–F. Price, 40 cts.

Surface Water Supply of Minor San Francisco Bay, Northern Pacific and Great Basins in California, 1895–1927. By H. D. MCGLASHAN. Pp. 68. U. S. Geol. Surv. Water Supply Paper 637–A, 1930. Price. 10 cts.

Foraminifera from the Atoka Formation of Oklahoma. By J. J. GALLOWAY AND C. RYNIKER. Pp. 37, pl. 5. Okla. Geol. Surv. Circ. 21, 1930 (Mar.). Systematic descriptions.

Sedimentation in the Anadarko Basin. By A. J. FREIE. Pp. 80, figs. 13, pl. 1. Okla. Geol. Surv. Bull. 48. Each formation is discussed under distribution, character, thickness, relations to adjacent formations, detailed sections and analyses, and origin.

Mica. By H. S. SPENCE. Pp. 142, figs. 10, pl. 21, maps 2. Canada Dept. of Mines No. 701, Ottawa, 1930 (Mar.). Price, 30 cts. Condensation of earlier publications; reviews the present status of the mica industry of Canada and of the world.

Horton-Windsor District, Nova Scotia. By W. A. BELL. Pp. 240, figs. 14, pl. (fossils) 36, map. Geol. Surv. of Canada Mem. 155, 1929 (Mar. 1930). Price, 50 cts. Structure, physiography, stratigraphy, sedimentation, and paleontology.

Indiana Oölitic Limestone: Relation of its Natural Features to its Commercial Grading. By G. F. LOUGHLIN. Pp. 90, figs. 10, pl. 18. U. S. Geol. Surv. Bull. 811–C, 1929 (Feb. 1930). Price, 30 cts. Geology, chemical properties, and grading.

The Rawlins, Shirley, and Seminoe Iron Ore Deposits, Carbon County, Wyoming. By T. S. LOVERING. Pp. 32, pl. 5, map. U. S. Geol. Surv. Bull. 811–D, 1929 (Mar. 1930). Geography, stratigraphy, structure, iron deposits.

Thirty-first Ann. Rept. of the Mining Industry of Idaho for the year 1929. By STEWART CAMPBELL. The annual report of the Inspector of Mines, giving a summary of the mineral occurrences of the State and the names, officers, capitalization, and locations of the various mining companies operating in Idaho mineral deposits.

Copies of books mentioned under "Reviews" or under our "New Book List" (see advertisement page) may be purchased through our Journal Bookshop by writing to W. S. Bayley, University of Illinois, Urbana, Ill.

SCIENTIFIC NOTES AND NEWS

John W. Gruner, associate professor in the Department of Geology and Mineralogy of the University of Minnesota, will investigate the origin of the iron ores of the Oliver Mining Company properties, under a gift of $1000 recently received from the Company for that purpose.

James D. Sisler has resigned his position as associate geologist of the State of Pennsylvania to become State Geologist of West Virginia.

Scott Turner, Director of the U. S. Bureau of Mines, represented the A. I. M. E. at the meeting of the Institution of Mining Engineers in Birmingham, England. He was also delegate from the United States to the World Power conference meeting in Berlin in June and the International Congress of Mining, Metallurgy, and Applied Geology at Liége the same month.

Carlton B. Hulin, of the University of California, spent a month recently in the Mojave Desert in work on the Searles Lake quadrangle for the State Division of Mines.

Arthur D. Storke, of the London office of the Rhodesian Selection Trust, has been on a trip to the copper properties in Northern Rhodesia, and later made a visit to New York City.

Herbert E. Munson, formerly geologist for the Texas Company in West Texas, has accepted the position of chief geologist for the Alberta-Pacific Consolidated Oil Company, with headquarters at Calgary, Canada.

N. H. Darton, of the U. S. Geological Survey, is spending the summer in work on the geological map of Texas, which is to be compiled this fall, and has requested that data on formation boundaries be sent in to him at Washington for use on this map.

James C. Templeton, of London, consulting geologist and geophysicist, recently made a geophysical survey of the properties of the British Controlled Oilfields, Ltd., in Venezuela.

W. G. Woolnough, Government Geologist for Australia, has been making an extended survey of oil fields in North and South America.

W. Thom, Jr., formerly of the U. S. Geological Survey, has taken up work under the American Association of Petroleum Geologists research project on the regional structure of Dakota sandstone in the Northern Plains region.

Bradford Willard, assistant professor of geology at Brown University, has resigned to take the position of associate geologist with the Topographic and Geological Survey of Pennsylvania.

A. C. Lawson, professor of Mineralogy and Geology at the University of California, is chairman of the program committee of the 1932 International Geological Congress, and is now in Europe on business connected with the program.

C. A. Farlow, formerly of the Miami Copper Company staff, has gone to the Urals on work for the U. S. S. R.

P. E. Peterson, consulting engineer of Vancouver, Canada, is now in Turkestan, and will spend two years in superintending silver-lead mining operations for the Soviet Government.

E. L. Bruce, of Queen's University, who has been studying pre-Cambrian formations in Finland, represented the Royal Society of Canada at the annual meeting of the Geological Society of France.

George Otis Smith was the speaker at the Commencement exercises of the Montana School of Mines. His subject was "The Engineer's larger Opportunity."

The Geological Society of America announces that the next annual meeting will take place in Toronto, Canada, Monday to Wednesday, December 29–31. The Paleontological and Mineralogical Societies, and probably the Society of Economic Geologists, will hold meetings at that time.

J. E. Brett, mining engineer and geologist of Toronto, Canada, was drowned on June 19, when the plane in which he was prospecting landed in rough water in the Albany river.

The recently published 20-volume index (336 pages) of ECONOMIC GEOLOGY for 1905 to 1925, compiled by J. M. Nickles, may be obtained for $3.00 from W. S. Bayley, University of Illinois, Urbana, Ill.

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